Abschlußbericht für das DLR-Vorhaben

Validation von SCIAMACHY level-2 Daten mit bodengebundenen DOAS-Messungen

Förderkennzeichen:

50 EE 0006

Laufzeit des Vorhabens:
01.09.2000 - 31.08.2004

Zuwendungsempfänger:
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Inhaltsverzeichnis

1. Zielsetzung des Vorhabens (Kurze Darstellung)

1.1 Aufgabenstellung und Voraussetzungen, unter denen das Vorhaben durchgeführt wurde

Bodengebundene MAXDOAS-Messungen sind aus verschiedenen Gründen besonders für die Validierung von Satellitendaten, insbesondere für die Messungen von SCIAMACHY, geeignet:
A) Der spektrale Bereich von DOAS Messungen deckt einen großen Teil des Bereichs von SCIAMACHY ab. Insbesondere können viele Spurengase in identischen spektralen Fenstern ausgewertet werden.
B) Zusätzlich zu den konventionellen Zenith-DOAS-Messungen werden im vorliegenden Projekt insbesondere auch MAXDOAS Messungen durchgeführt. Diese erlauben die Bestimmung der partiellen Säulen (in der Troposphäre und der Stratosphäre) vieler Spurengase. Die Validation nicht nur von atmosphärischen Gesamtsäulen, sondern von troposphärischen und stratosphärischen Teilsäulen verschiedener Spurenstoffe ist von besonderer Bedeutung für SCIAMACHY, da der gekoppelte Limb/Nadir-Modus speziell die Bestimmung von Teilsäulen erlaubt.
C) Die für die Validation vorgesehenen Bodenstationen erstrecken sich über einen weiten Bereich der geographischen Breite: Außer den polaren Stationen in Kiruna, Neumayer und Arrival Heights wurde im Rahmen dieses Projektes auch ein Meßgerät in den Tropen (Paramaribo, Surinam) aufgebaut. Somit lassen sich breitenabhängige (also z.B. auch Sonnenzenithwinkelabhängige) Effekte von SCIAMACHY detailliert untersuchen. Die Messungen in Paramaribo sind die ersten Dauermessungen eines bodengebundenen MAXDOAS-Systems in den Tropen.

1.2 Planung und Ablauf des Vorhabens und Wiss. und techn. Stand zu Beginn des Projektes

Das Institut für Umweltphysik der Universität Heidelberg besitzt langjährige Erfahrungen mit bodengebundenen DOAS-Messungen [Fiedler et al., 1990; Erle et al., 1995; Platt et al., 1997; Otten et al., 1998; Wagner et al., 1998; 2000; 2002; 2004; Frieß et al., 1999; 2001; 2004; 2005]. Für die geplanten Validationsaufgaben mußte ein neues MAXDOAS-Meßinstrument konstruiert und auf der Station von Paramaribo (Surinam) in Betrieb genommen werden. Zusätzlich wurde das Zenith-Gerät auf der Station Neumayer für MAXDOAS-Messungen umgerüstet.
Dadurch wurde es möglich, durch diese Messungen detaillierte Messungen für troposhärische Teilsäulen durchzuführen.
Der Neu- und Umbau der Geräte bestimmten die Tätigkeiten während des ersten Jahres. In den folgenden Jahren wurden die Geräte betreut und der Transfer der Meßdaten (z.T. per Post, später auch über das Internet) nach Heidelberg organisiert. Die Datenanalyse wurde anhand neuester Ergebnisse, und in enger Kooperation mit den DOAS-Gruppen in Bremen und Brüssel, durchgeführt. Ein Hauptaugenmerk lag dabei auf einer konsistenten Behandlung der verschiedenen Meßreihen.
Speziell für das Gerät in Kiruna waren mehrere Besuche zur Reparatur und zum Ersatz eines Spektrographen nötig. Bis auf die damit verbundenen Unterbrechungen waren alle Meßgeräte kontinuierlich im Einsatz.

1.3 Zusammenarbeit mit anderen Stellen

Eine besonders enge Kooperation bestand mit der DOAS-Gruppe der Universität Bremen. Grundsätzliche instrumentelle Entwicklungen und Analysemethoden wurden gemeinsam vorangetrieben. Hierbei bestand auch ein enger Austausch mit der DOAS-Gruppe von IASB/BIRA in Brüssel. Die Messungen der Station in Surinam sind in das Europäische Forschungsprojekt (STAR, http://www.knmi.nl/samenw/star/station.html) integriert; mit den Projektteilnehmern besteht eine enge Zusammenarbeit. Die Station Neumayer gehört dem internationalen NDSC-Netzwerk an.

2 Eingehende Darstellung

Für die eingehende Darstellung wurde die englische Sprache gewählt. Gerade im Bereich der Satelliten-Datenanalyse besteht eine enge internationale Zusammenarbeit. Moderne optische Sensoren und Analysemethoden für atmosphärische Spurenstoffmessungen erleben gerade eine starke weltweite Verbreitung.
Der vorliegende Bericht beinhaltet viele Details zu Instrument und Auswertemethoden, die für viele internationale Gruppen von Interesse sind.
Die Zusammenfassung der Ergebnisse (Abschnitt 2.6) ist in deutscher Sprache verfasst.

2.1 General Motivation of the project

Within this project comprehensive validation products from ground based DOAS observations are created. The most important advantage of such data sets is their great similarity to the data products of SCIAMACHY. In particular, many so called weak absorbers like e.g. BrO can be measured by ground based DOAS instruments. In addition, the measurement locations cover very different latitudes; thus the performance of SCIAMACHY in different climatic zones, especially with respect to the dependence on the solar zenith angle can be studied.

2.2 Overview on the different stations

In Fig. 1 an overview on the Heidelberg network of ground based DOAS stations is shown. While the polar stations at Kiruna, Neumayer, and Arrival Heights were already in operation at the beginning of this project, the instrument for the new station at Paramaribo (Suriname) was build and installed within the project.
The stations and the geographical coordinates are:

- Kiruna (Sweden, 67.84° N, 20.41° E)
- Paramaribo (Suriname, 6°N, 55°W)
- Neumayer (Antarctica, 70.6°S, 8.3°W)
- Arrival Heights (Antarctica, 77.8°S, 166.7°E)

The periods of operation of the different instruments is shown in Fig. 2.

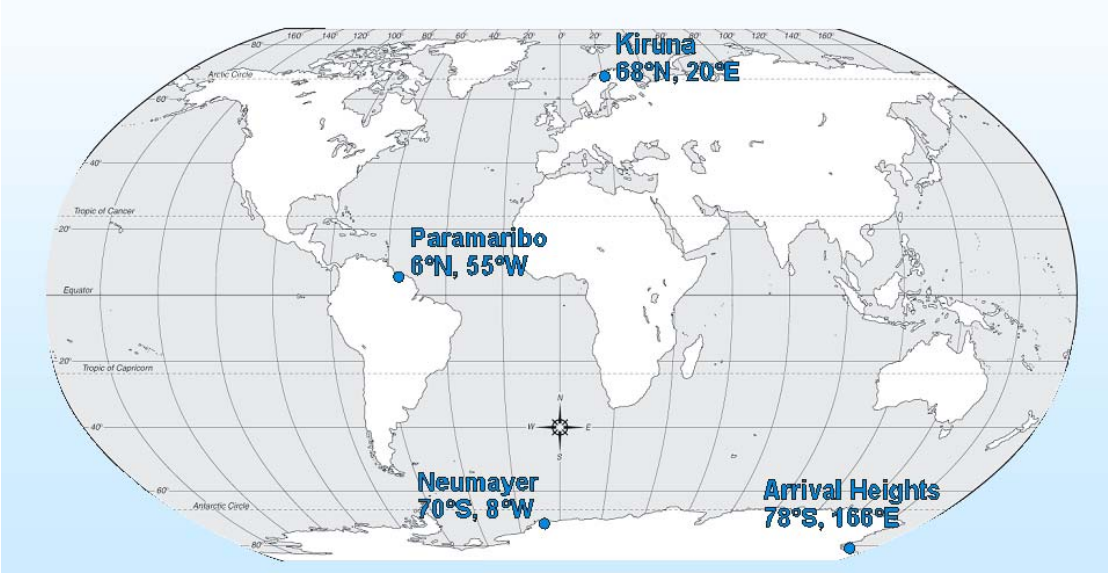

Fig. 1 Overview on the Heidelberg network of ground based DOAS stations. The instrument at Paramaribo (Suriname) was build and installed within the project.

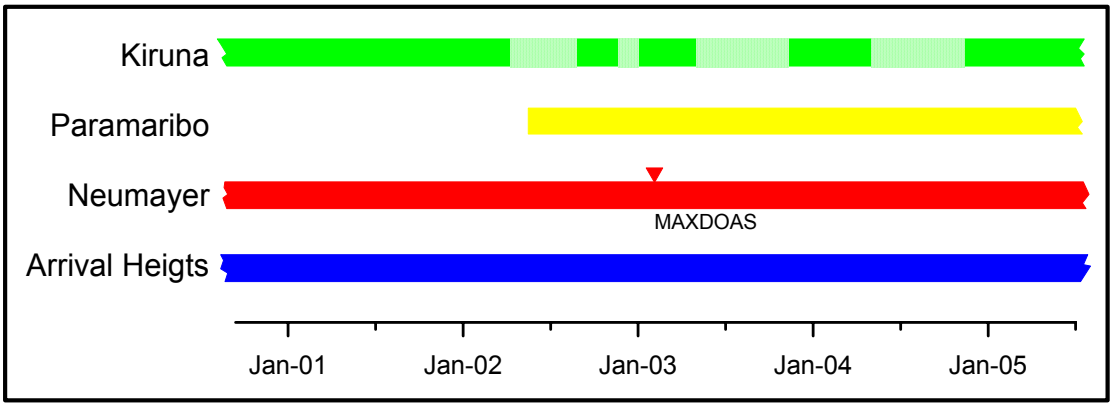

Fig. 2 Periods of successful measurements of the different stations. The Paramaribo measurements started in May 2002. In early 2003 the instrument at the Neumayer station was equipped with a MAXDOAS telescope. The light green colour indicates times when the Kiruna measurements were at part of the Kiruna instruments were not in operation (see section 2.2.1).

2.2.1 Kiruna (Sweden, 67.84° N, 20.41° E)

The ground based DOAS measurements at the Institute of Space Physics (Institutet för rymdfysik, IRF) in Kiruna started at December 1996 (http://www.irf.se/program/afp/doas/). The instrumental set-up consists of three instruments:

1. An UV spectrometer, in operation since December 1996 - present
2. A vis spectrometer, in operation since December 1996 - 2003.

A detailed description of both instruments is given in Otten et al., [1998] and Roscoe et al., [1999].
3. A second vis spectrometer, since May 2002 - present.
A more detailed description of this new instrument can be found in Bossmeyer, [2002] and Hönninger et al., [2002].

The instruments are installed in the optics laboratory of the IRF on an elevated table directly below a plexi-glass dome (see Figures 3 and 4). The room temperature is stabilised and the dome is ventilated with air to prevent condensation and freezing of water vapor. All three instruments are controlled via one computer, which (together with the various instrument controllers) are installed below the instrument table. The three telescope lenses are mounted on a common frame over which a black shielding or a halogen lamp is automatically moved during night (see Fig. 5). These nightime measurements are important for the calibration of the instrument and the correction of dark current and electronic offset. In May 2002, a new visible spectrometer was installed (Fig. 6) after the existing visible instrument showed strong and increasing spectral structures which caused problems for the spectral analysis.

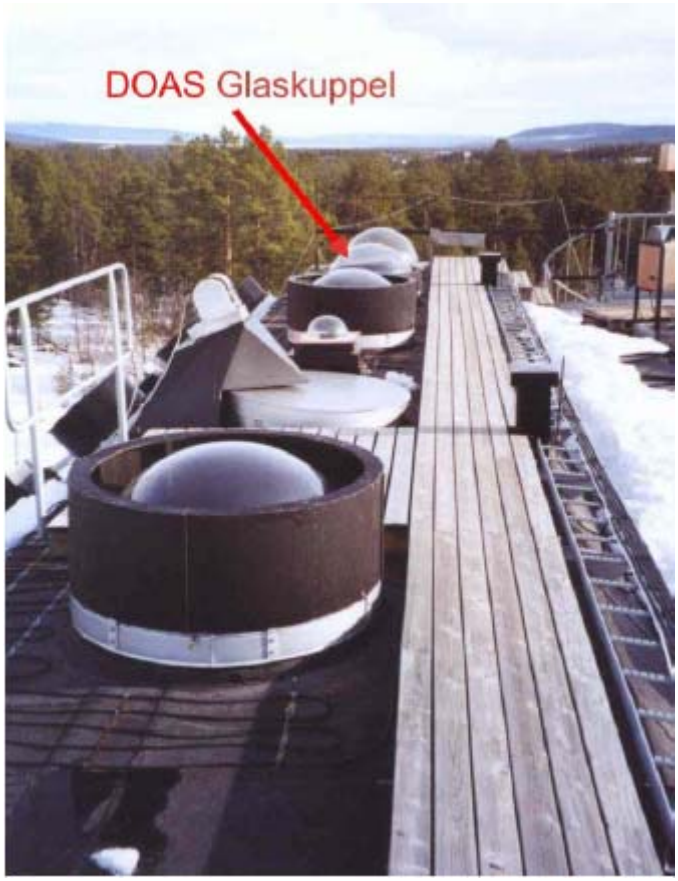

Fig. 3 View on the roof of the Institute of Space Physics (Institutet för rymdfysik, IRF) in Kiruna (Sweden). Below the plexi-glass dome indicated by the arrow is the optical laboratory hosting the Heidelberg DOAS instruments.

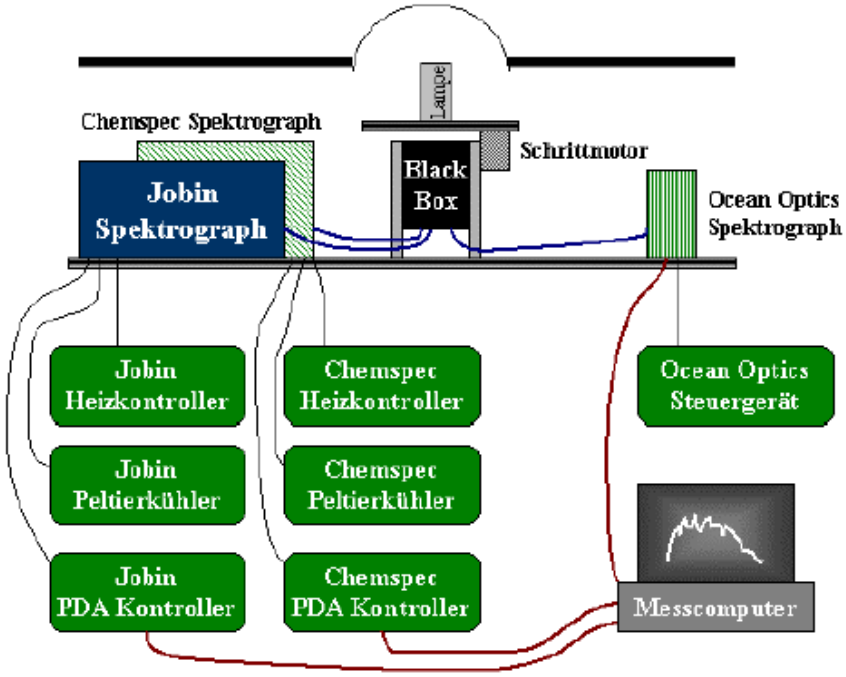

Fig. 4 Instrumental set-up of the Kiruna instruments. The three spectrometers are mounted on a high table directly below the plexi-glass dome. The computer and the controlling devices are placed below.

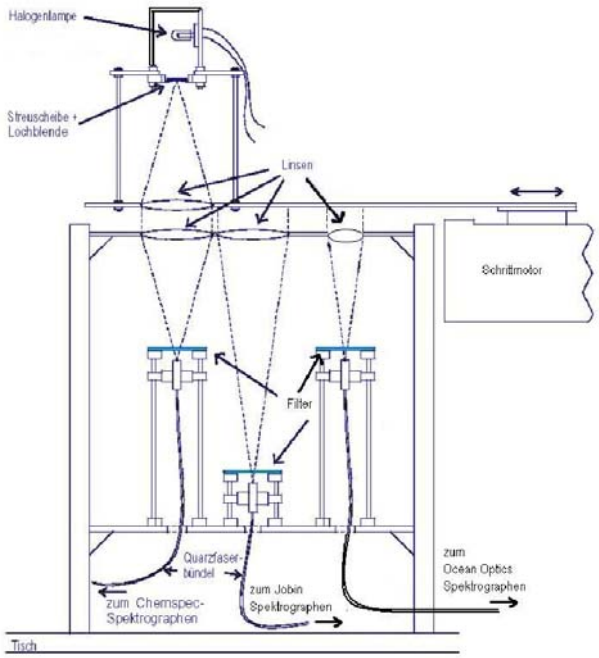

Fig. 5 The telescope lenses for the three spectrometers are mounted on a common frame over which a black shielding or a halogen lamp is automatically moved during night. These measurements are important for the calibration of the instrument and the correction of dark current and electronic offset (Bugarski, 2003).

Fig. 6 New visible spectrometer installed in May 2002. Its telescope optics was integrated into the existing telescope set-up (see Fig. 5).

Unfortunately, especially in the period after the launch of SCIAMACHY, the Kiruna instrument experienced repeated problems of the computer controlling. Because of the old age of the UV-instrument, it was difficult to find compatible replacements for parts which stopped working properly. Eventually, it was always possible to restart routine operation of the instruments again. However, in several cases this was only possible after personal visits at the instrument, which caused several longer gaps in the data series (see Fig. 2). It is important to note here that the Kiruna instrument is one of the instruments with the longest measurement history in the Arctic. Continuous data sets without discontinuities are of high values especially for trend analysis of stratospheric constituents.

2.2.2 Paramaribo (Suriname, 6°N, 55°W)

In May 2002, two MAXDOAS instruments, one for the UV, and one for the visible spectral range were installed at the meteorological service of Paramaribo, Suriname (See Fig. 7). Paramaribo is located at 6°N (see Fig. 8), which is very important for SCIAMACHY validation, since only very few ground based stations exist in the Tropics. At the time of installation, the MAXDOAS instrument at Paramaribo was – to our knowledge – the first continuously operating DOAS instrument in the Tropics.

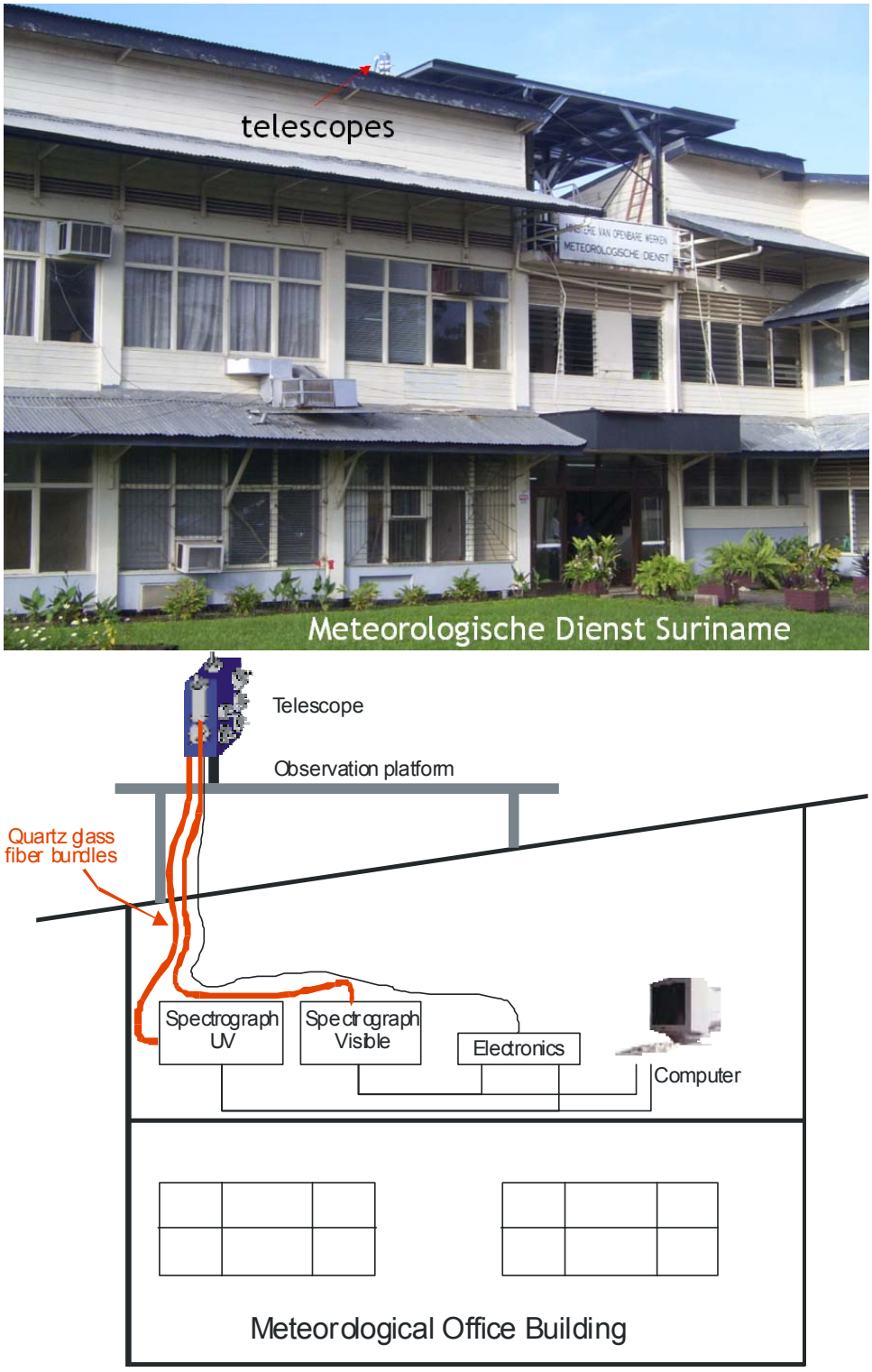

Fig. 7 Top: Building of the meteorological service of Suriname in Paramaribo. The telescopes of the MAXDOAS instrument are seen at the top (see also Fig. 9). Bottom: The telescope units outside the building are connected to the spectrometers inside via glass fibre bundles.

Fig. 8 Map of Suriname. The meteorological station is located in the capital, Paramaribo.

back view: southeast over bush

front view:northwest over city

Fig. 9 telescope units or the UV and visible spectrometer at Paramaribo. The static telescopes mounted at the blue housing belong to the UV instrument; the separate telescope at the side of the housing belongs to the visible instrument and is moveable via a stepper motor.

Fig. 10 Indoor set-up of the MAXDOAS instruments. The aluminium box at the bottom hosts the UV spectrometer; the small silver device at the right side of the board is the visible spectrometer.

Two instruments have been set-up:
The VIS light is fed into an Ocean Optics USB 2000 spectrograph that disperses the light and maps it onto a one dimensional CCD array with 2048 elements (see Fig. 11). The vis-spectrograph was mounted into a modified hermetic Dewar vessel. In order to avoid ambient air from penetrating into the protective vessel it had been evacuated and filled up with gaseous Argon up to 1.2bar prior to the expedition. Stable measurement conditions and low detector noise could be achieved by cooling the spectrograph down to ca. –15°C by a two-stage Peltier cooling unit. A radiator with fan served as a lid for the Dewar, the fan blowing away the heat created in the cooling process. The electronic signal that was produced in the detector array by the incoming light was sent via an integrated analogue to digital converter (ADC) to a read-out unit of a computer, which stored and imaged the spectra.
The UV light, on the other hand, was fed into an Acton Spectra Pro 300i spectrograph that dispersed the light and mapped it onto a two dimensional CCD camera manufactured by Andor (model DV 420-OE) with 256 vertical times 1024 horizontal pixels (see Fig. 12).
The CCD-camera was put into a modified and insulated aluminium box. Since the UV measurements of the different viewing directions were carried out simultaneously the light of each optical fibre, two fibres for every line of sight, was arranged on a

different assembly of detector rows. The stability of the set-up was ensured by cooling the camera to –28°C and heating the spectrograph to +30°C.
The Suriname MAXDOAS measurements are continuously in operation since May 2002.

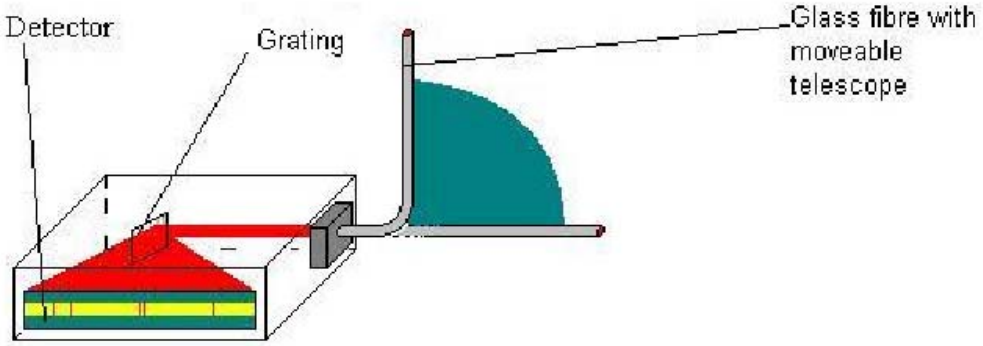

Fig. 11 Scheme of spectrograph and detector arrangement for the visible spectral range. The light is dispersed onto an one-dimensional CCD-detector.

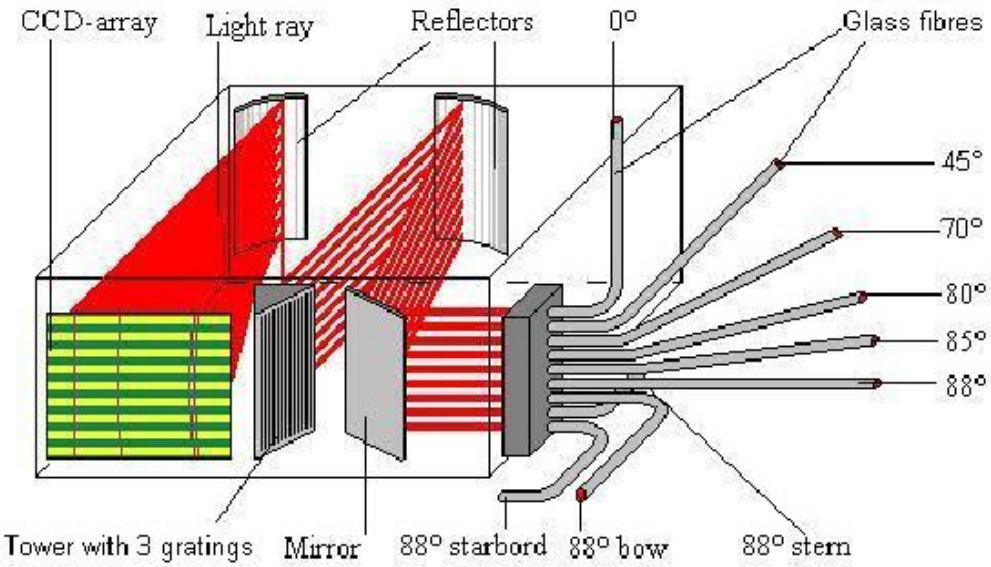

Fig. 12 Scheme of spectrograph and detector arrangement for the UV. The light from the different telescopes is fed into the spectrometer at different vertical positions corresponding to different spectra on the two-dimensional CCD-detector.

2.2.3 Neumayer (Antarctica, 70.6°S, 8.3°W)

The DOAS instrument at the German Antarctic research station Neumayer has been installed in cooperation with the Alfred Wegener Institute for Polar and Marine Research (AWI, Bremerhaven/Germany) in 1999. The instruments were developed and built on the basis of a DOAS instrument for balloon-borne measurements [*Ferlemann et al.,* 2000; *Frieß,* 2001]. In brief, it consists of a dual-channel spectrograph/detector system covering the wavelength region of 320 - 640 nm. The whole spectrograph/detector system is located in a pressurised and temperature stabilised stainless steel vessel, ensuring an operation under very stable conditions – an important prerequisite for long-term measurements of high quality (Fig. 13).
The Neumayer instrument was initially equipped with a zenith sky telescope only. A new multi-axis telescope has been developed by Dr. Udo Frieß and installed in early 2003 (Fig. 14), allowing to observe scattered sunlight from any elevation angle using rotating quartz glass prisms.
The ground based measurements are continuously in operation since 1999.

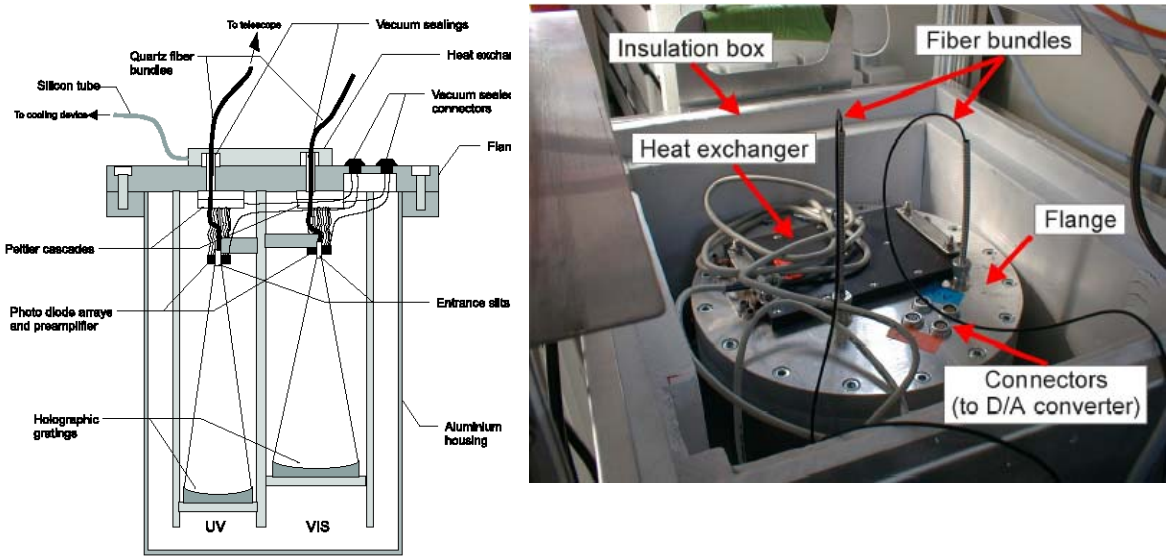

Fig. 13 Left: Scheme of the two DOAS spectrometers at the Neumayer station [Frieß, 2003]. They are integrated together inside a pressure and temperature stabilised housing. Right: Picture of the top of the spectrometer housing showing the various connections to the telescopes and the controlling devices.

Fig. 14 New MAXDOAS telescope installed in early 2003. The Multi-Axis-Observations allow to infer more detailed information on tropospheric trace gases.

2.2.4 Arrival Heights (Antarctica, 77.8°S, 166.7°E)

The MAXDOAS instrument at the station of Arrival Heights was installed in 1998. It is operated by Dr. Karin Kreher from NIWA/New Zealand, which has agreed to provide the analysed data for SCIAMACHY validation. The instrument was developed and built by Dr. Udo Frieß; it is almost identical to that installed at the Neumayer station. The entrance optics is equipped with a tracker system developed by NIWA. Mounted on the roof of the Arrival Heights observatory, it allows to observe scattered sunlight from any direction of the sky using two moveable mirrors (see Figure 15). More details of the instrument can be found in Kreher et al., 2002.

Figure 15 The tracker system of the Arrival Heights MAX-DOAS instrument.

2.3 Measurement principle and radiative transport calculations

The Multi AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) technique allows to separate the absorptions taken place at different altitudes in the atmosphere by observing scattered sun light from a variety of viewing directions (see Fig. 16) [Hönninger and Platt, 2002; Von Friedeburg et al., 2003; Bobrowski et al., 2003; van Roozendael et al., 2003; Wittrock et al., 2003; Hönninger et al., 2004; Heckel et al., 2004; Wagner et al., 2004]. This is possible because for most measurement conditions, the observed light is scattered in the free troposphere. Thus air masses located close to the ground are traversed on a slant absorption path determined by the viewing direction; in contrast, stratospheric air masses are traversed on a slant absorption path determined by solar zenith angle (see Fig. 17, 18).
An overview on the different trace gas products is shown in Table 1.
In addition to the partial columns of various trace gases (like NO_2, BrO, HCHO, etc.) also information on the aerosol profile can be retrieved [Wagner et al., 2004].

Table 1 Data products of the MAXDOAS measurements. For most of the trace gases parts of the total column are located in the stratosphere and the troposphere. From MAXDOAS observations both partial columns can be discriminated.

Trace gas	**Spectral range**	**Tropopsheric and/or stratospheric columns**
O_3	UV / vis	Strat/trop
NO_2	UV / vis	Strat/trop
BrO	UV	Strat/trop
OClO	UV / vis	Strat
H_2O	vis	Trop
HCHO	UV	Trop
O_4	UV / vis	Trop
IO	UV / vis	Strat?/trop

The sensitivity of DOAS observations of scattered light is usually quantified by the so called air mass factor, which is the ratio of the trace gas concentration integrated along the light path (SCD) and the vertically integrated trace gas concentration (VCD).

AMF = SCD/VCD

It is usually determined from numerical radiative transfer calculations [Solomon et al., 1987; Marquard et al., 2000]. According to the altitude of a trace gas in the atmosphere, the respective AMF depends strongly on the SZA or on the elevation angle (see Fig. 17, 18).
For the analysis of the MAXDOAS observations within this project a new radiative transport model (TRACY) was developed, which is based on the backward Monte Carlo method [vonFriedeburg, 2003; vonFriedeburg, 2003; Wagner et al., 2004]. It was explicitly designed for the correct treatment of spatial gradients, which is important for the correct interpretation of MAXDOAS observations.
In addition to the retrieval of concentration profiles of the atmospheric trace gases, also profiles of aerosol properties (like extinction, single scattering albedo, and phase

function) can be analysed from MAXDOAS of the oxygen dimer O_4 [Wagner et al., 2004; Sinreich et al., 2005].

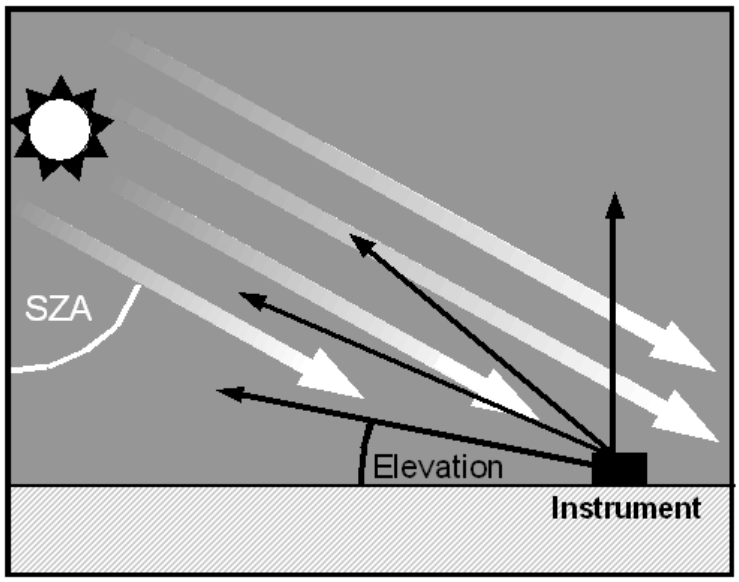

Fig. 16 Scheme of the MAX-DOAS observation geometry. Scattered sunlight is observed under various elevation (and often also azimuth) angles. Often the elevation angle of the telescopes is defined with respect to the ground. The solar zenith angle (SZA) is defined as the angle between the incident sunlight and the zenith.

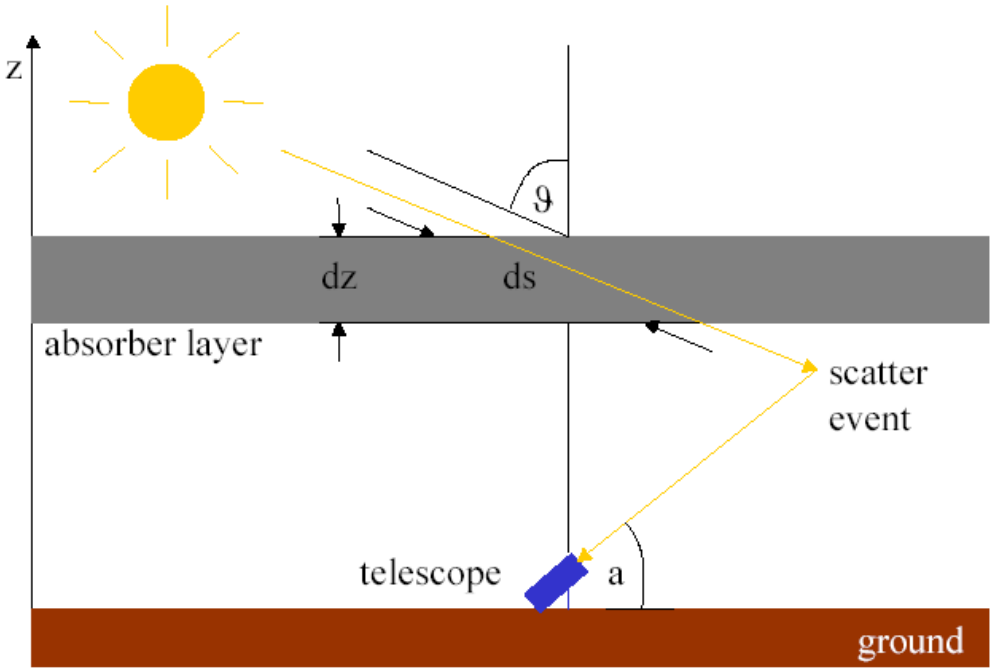

Fig. 17 The path length through the stratosphere is mainly determined by the solar zenith angle (SZA); from observations made at different SZA, the stratospheric trace gas absorption can be retrieved.

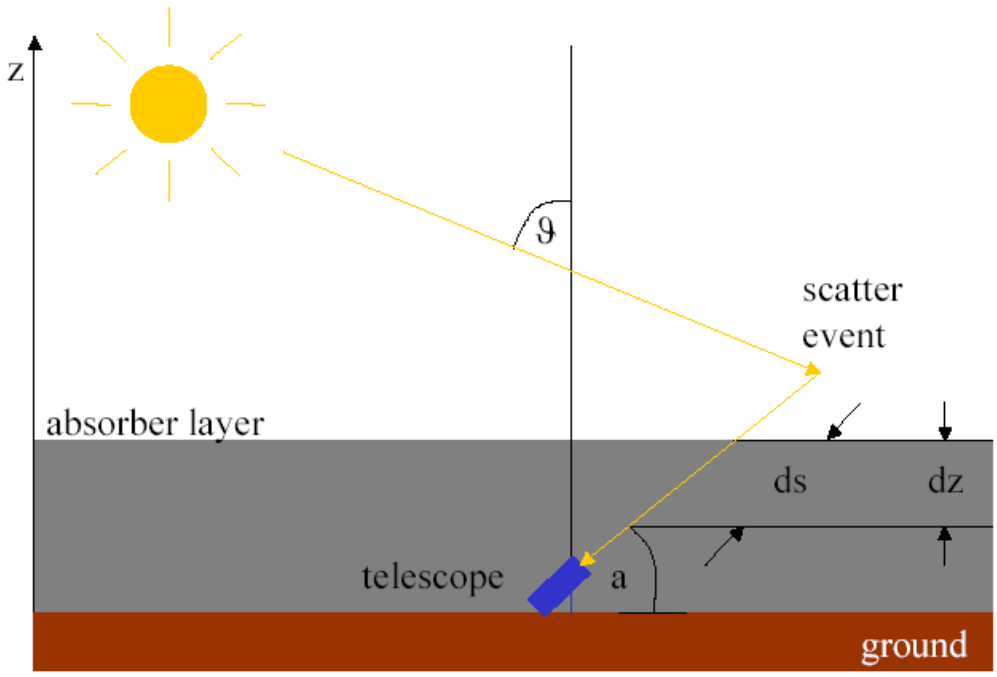

Fig. 18 The path length through the boundary layer is mainly determined by the viewing angle of the telescopes.; from observations made at different viewing angles, the boundary layer trace gas absorption can be retrieved.

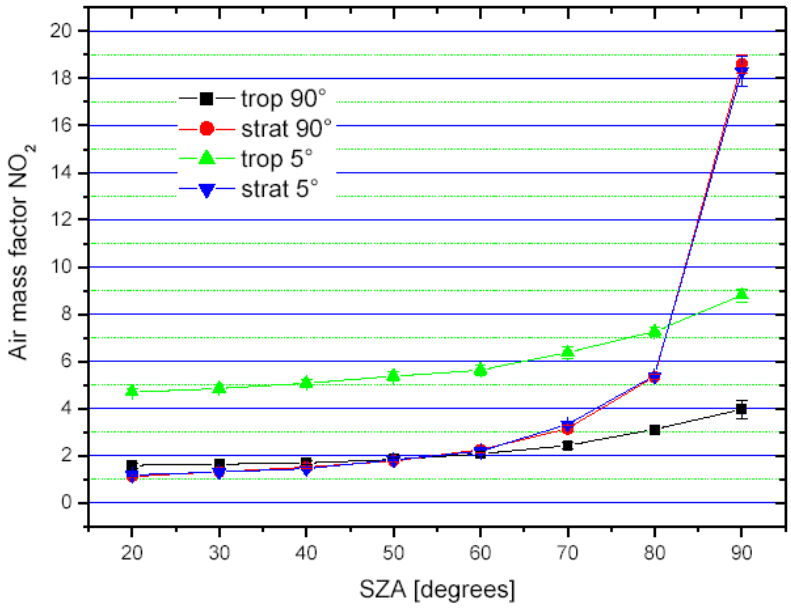

Fig. 19 Tropospheric and stratospheric air mass factors of NO_2 for elevation angles 5° and 90° calculated with the Heidelberg radiative transfer model TRACY. The AMFs for stratospheric concentrations mainly depend on the SZA; the AMFs for stratospheric concentrations mainly depend on the elevation angle.

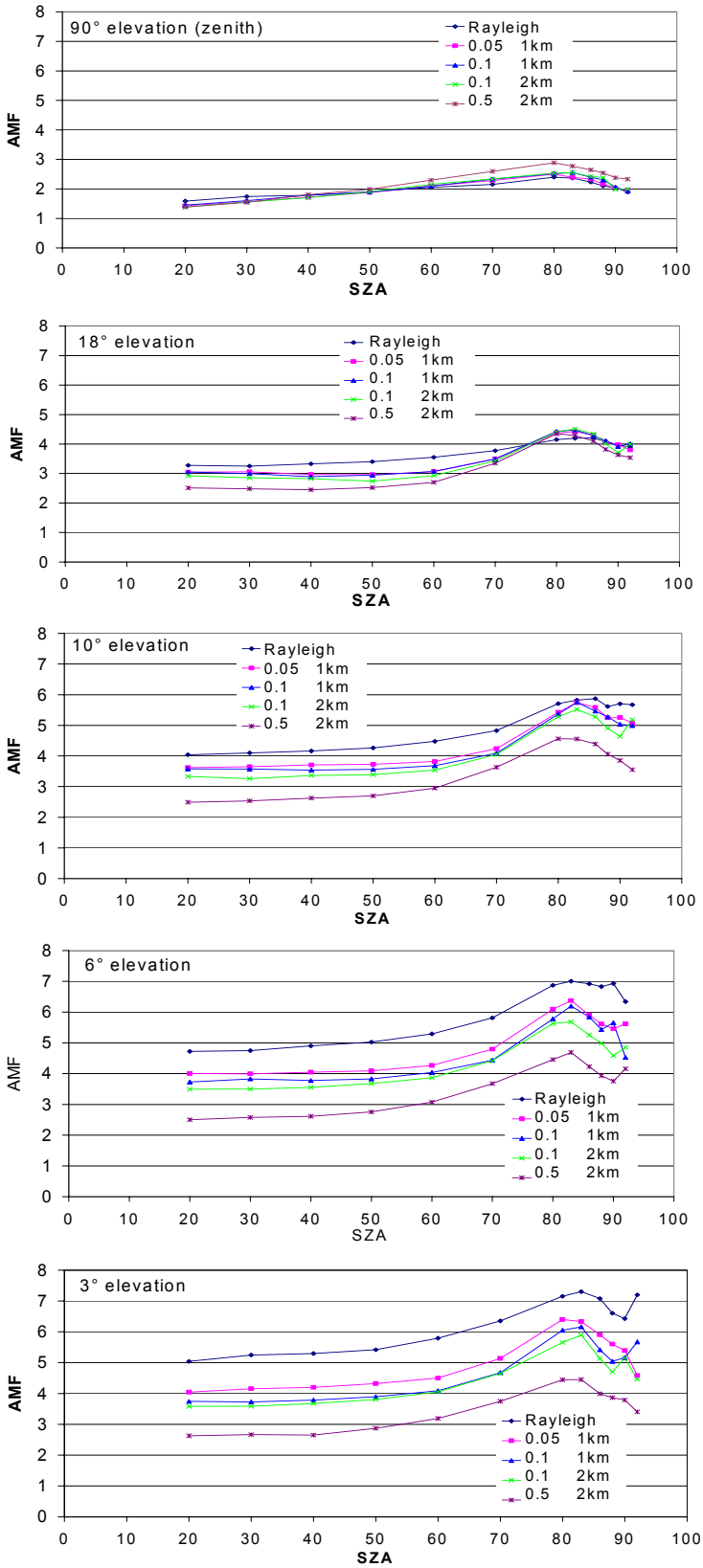

Fig. 20 O_4 AMF calculated with the Monte Carlo model 'TRACY' [Von Friedeburg, 2003; Wagner et al., 2004] for different MAX-DOAS elevation angles and aerosol scenarios. The first number in the legend indicates the aerosol extinction per km, the second number the upper boundary of the aerosol layer.

In Figures 19 characteristic AMFs for tropospheric and stratospheric AMFs are shown. It becomes clear that the stratospheric AMFs mainly depend on the solar zenith angle, whereas the tropospheric AMFs mainly depend on the elevation angle.
Figure 20 shows additional calculations of tropospheric AMFs of O_4 under different conditions of aerosol scattering [Wagner et al., 2004]. MAXDOAS O_4 observations are very sensitive even to very small atmospheric aerosol extinctions [Wagner et al., 2004; Sinreich et al., 2005]

2.4 Spectral analysis

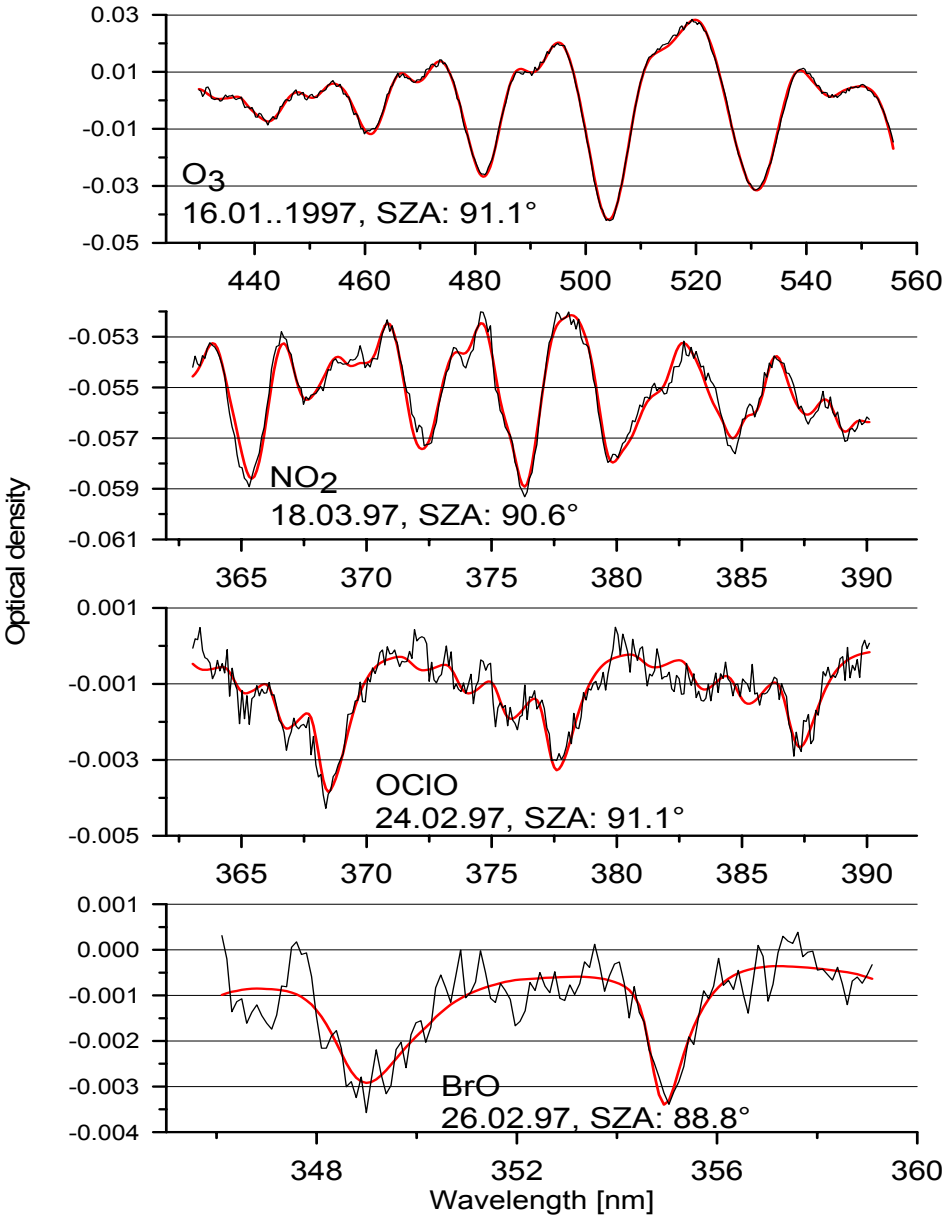

Fig. 21 Examples of the spectral fitting procedure of the different trace gases (data from the Kiruna instrument). Displayed are the absorption cross sections (red curves) scaled to the respective trace gas absorption in the measured spectrum (black curves).

The measured spectra are analysed using the DOAS method [Platt, 1994]. To the (logarithm of the) measured spectrum several trace gas cross sections as well as a Ring spectrum [Grainger and Ring, 1962; Bussemer, 1993], a Fraunhofer reference spectrum and a polynomial of low degree are fitted by means of a least squares fitting routine [Gomer et al., 1993; Stutz and Platt, 1996]. Also included in the fitting routine was an inverse Fraunhofer spectrum ($1/I_0$) for the correction of a possibly remaining intensity offset. The wavelength calibration was performed by fitting the measured spectra to a high resolution solar spectrum [Kurucz et al., 1984]. Examples of the DOAS analysis of the various trace gases are shown in Fig. 21.

2.5 Results and comparison with satellite data

Because of the very extensive data sets derived from the different ground based stations, it was so far not possible to analyse the stratospheric and tropospheric columns of all trace gases of interest. According to the quality and availability of the different trace gas products from SCIAMACHY, the first validation activities focussed on the total atmospheric columns (in particular the total VCDs of O_3, NO_2 and BrO).
In addition, also partial columns for the troposphere were retrieved, in particular for NO_2, H_2O, and O_4. Due to the lack of respective SCIAMACHY data products, these data could so far not been used for the satellite validation.
Unfortunately, we only received little data from our New Zealand colleagues for the station of Arrival Heights. However, a comprehensive validation study was possible for the station Neumayer, which is well representative for Antarctica.

2.5.1 Stratospheric O_3

The total atmospheric column of ozone was analysed for the stations of Kiruna, Paramaribo, and Neumayer (Figures 22 – 24).
In general, very good agreement between the ground based and satellite measurements was found. The largest differences were found for Paramaribo; the reasons are not fully understood yet but the apparent drift of the ground based time series indicates a potential problem in the analysis of the MAXDOAS data.
In addition to the absolute values, also the correlation between the MAXDOAS and satellite observations was investigated. For this purpose, the Neumayer data have been chosen, because for the polar conditions the strongest variations have been observed. The slope was found to be 0.916, indicating the good agreement between both data sets. Remaining differences might be at least partly be related to temporal and spatial variations close to the vortex boundary.

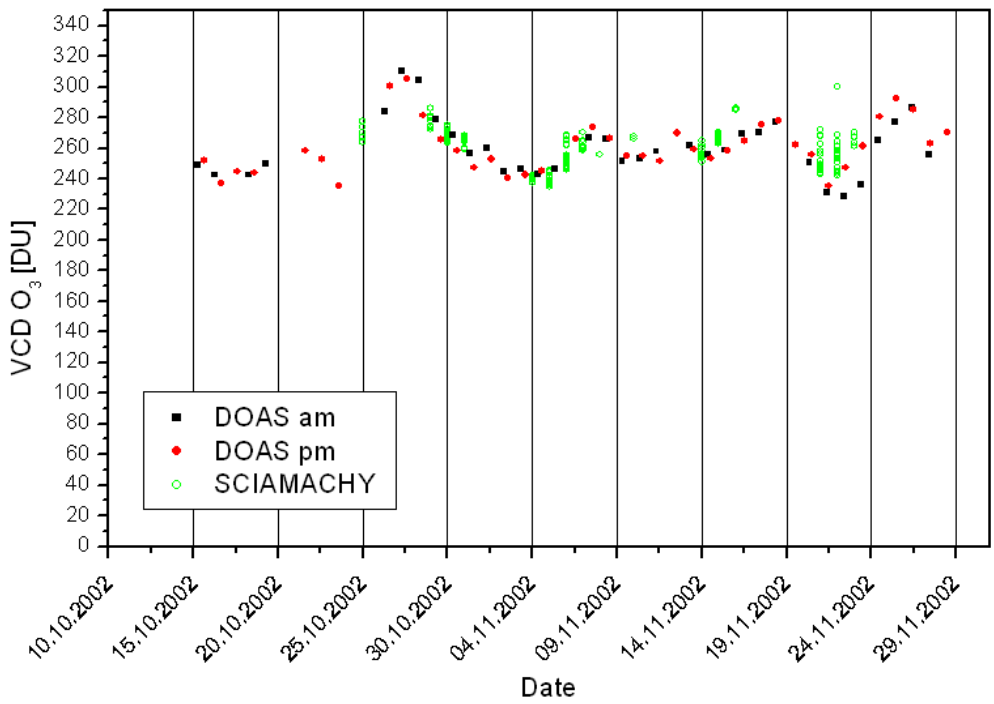

Fig. 22 Comparison between the total columns of O_3 from the ground based measurements at Kiruna and those from SCIAMACHY.

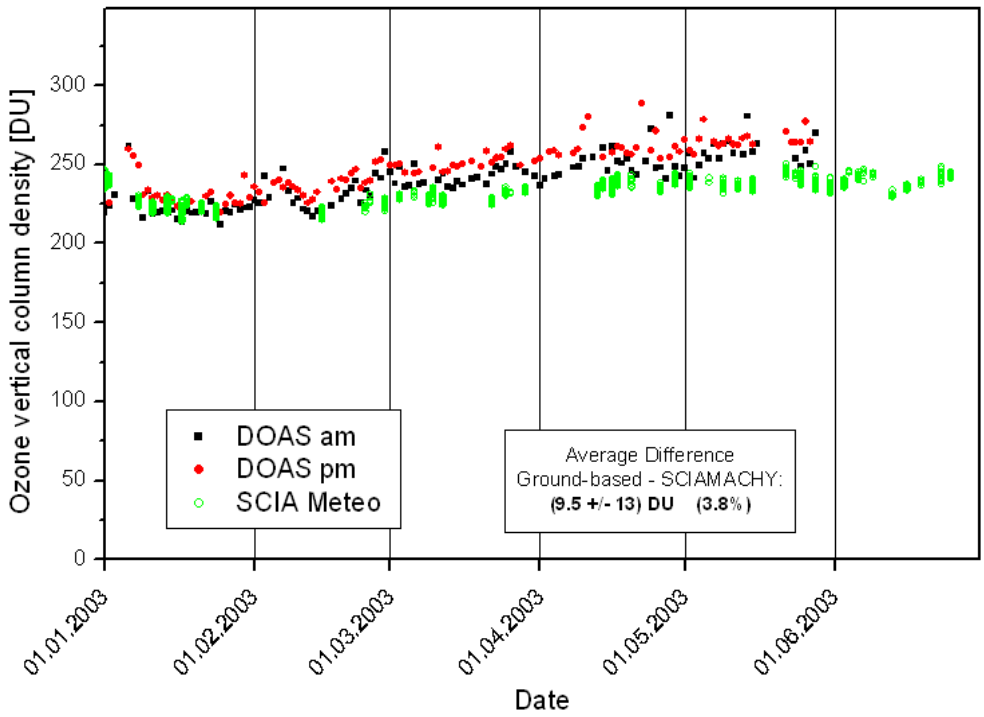

Fig. 23 Comparison between the total columns of O_3 from the ground based measurements at Paramaribo and those from SCIAMACHY.

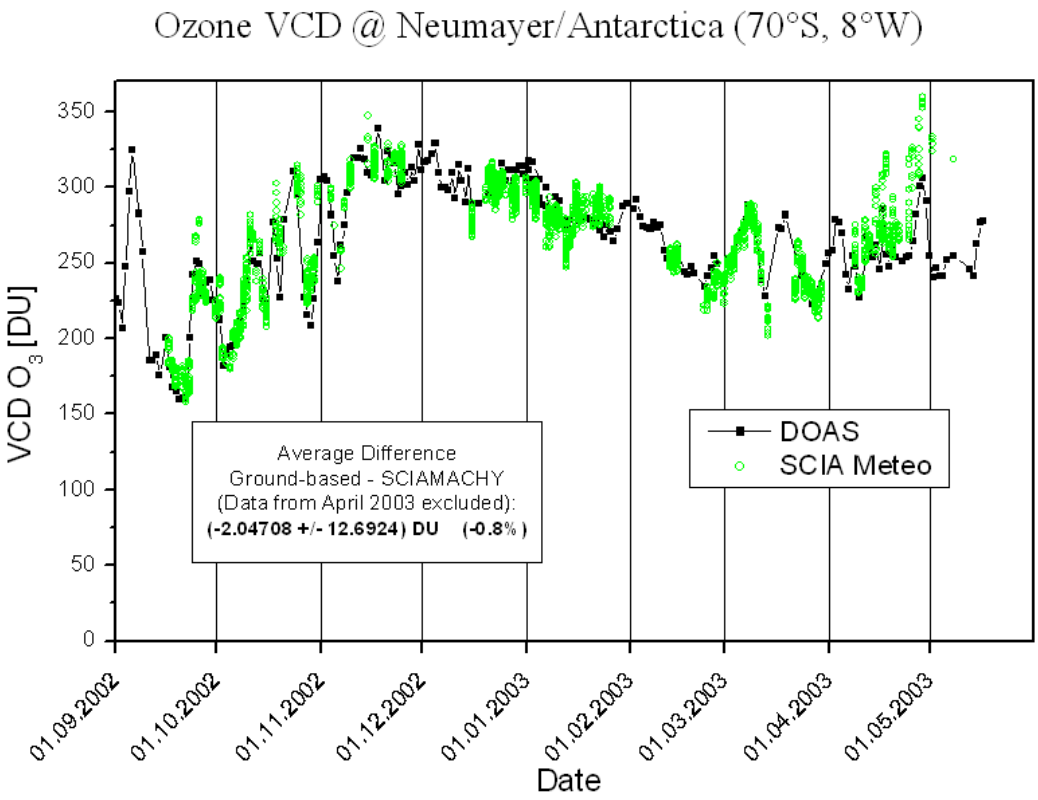

Fig. 24 Comparison of the total columns of O_3 from the ground based measurements at Neumayer and those from SCIAMACHY.

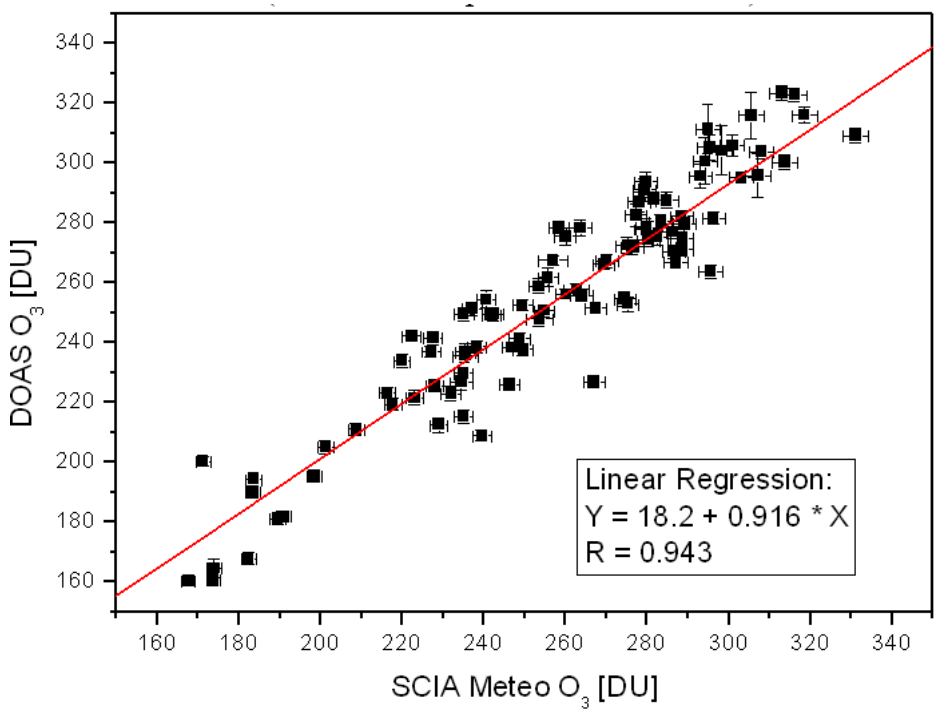

Fig. 25 Correlation analysis between the total columns of O_3 from the ground based measurements at Neumayer and those from SCIAMACHY.

2.5.2 Stratospheric NO_2

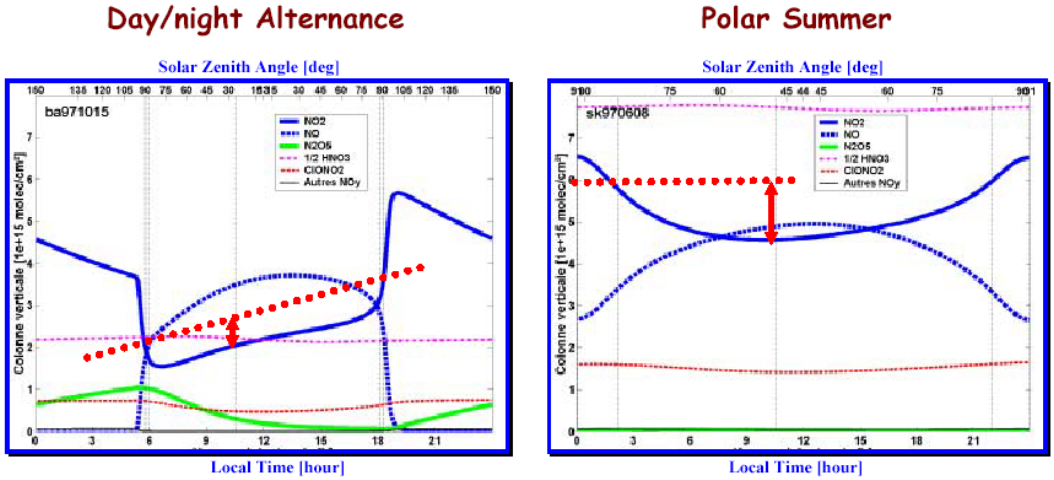

Fig. 26 Diurnal cycle of reactive nitrogen compounds for two selected regimes. Left: for most observations N_2O_5 is accumulated during night and photolised during day. Under these conditions the NO_2 VCD during sunrise is very similar to that around noon when the satellite overpass occurs. The NO_2 VCD during sunset is, however, much larger. Right: during polar summer, only the fast photolysis of NO_2 determines the diurnal cycle of NO_2. The NO_2 VCD during sunrise and sunset is systematically larger than at noon [Lambert et al., 2002].

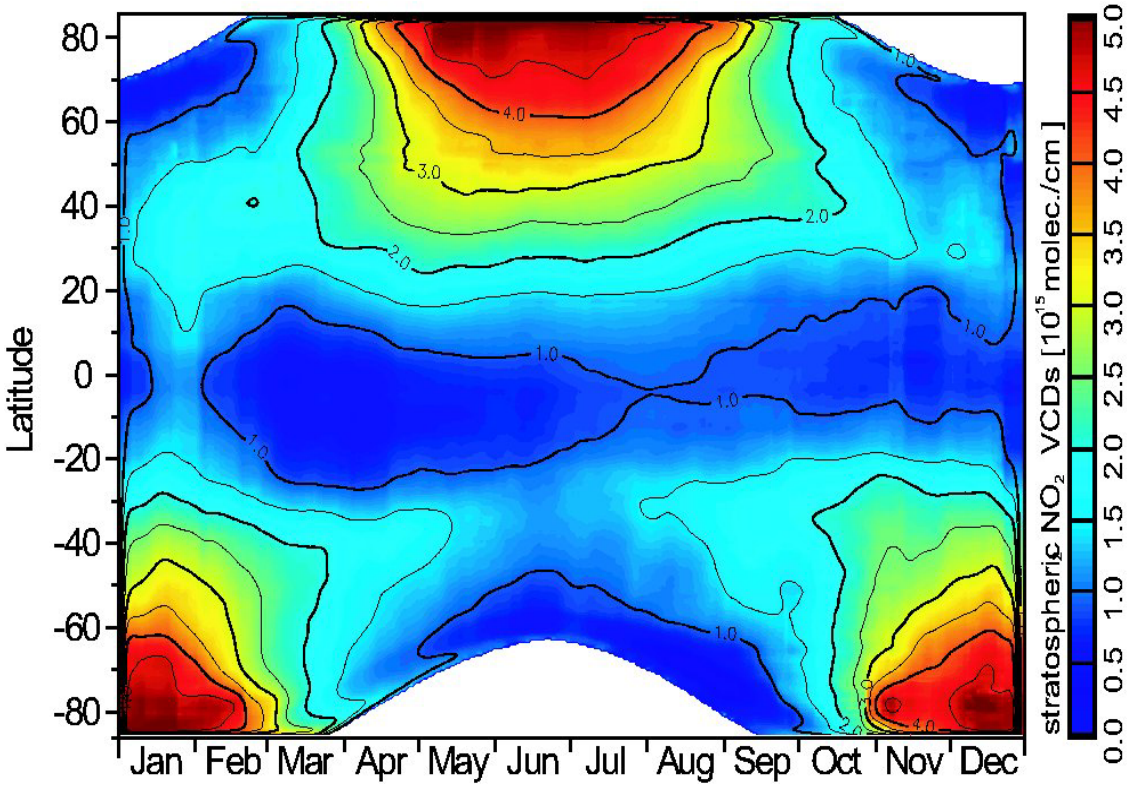

Fig 27 Mean annual cycle (1996 - 2001) of the stratospheric NO_2 VCD analysed from GOME observations. Each pixel represents zonal mean values. The contour lines are in units of 10^{15} molecules/cm^2 [Wenig et al., 2004].

For NO_2 so far the stratospheric VCD was the major target quantity for satellite validation; the stratospheric VCDs is typically analysed at high solar zenith angle.
Thus, for the comparison to SCIAMACHY (and other satellite) data two aspects have to be taken into consideration. First, in contrast to the ground based observations made at high solar zenith angle, the satellite observations are usually performed for low SZA; thus their sensitivity towards tropospheric contributions is in general much higher. If substantial tropospheric NO_2 concentrations are present, the total NO_2 VCDs retrieved from the satellite instruments are thus systematically higher than those analysed from MAXDOAS observations. Second, because of the photochemistry of NO_2, the stratospheric NO_2 concentrations during noon (when the satellite takes the measurements) are typically much lower than during sunset. The observations during sunrise are, however, in many cases very similar to the satellite observations, because the effects of photochemical decomposition of N_2O_5 and of NO_2 both almost compensate each other (see Fig. 26).
Besides the stratospheric temperature, in particular the availability of sun light determines the NO_2 concentration. During polar night, the smallest concentrations occur; during polar day the highest concentrations are found (see Figures 27 -29).

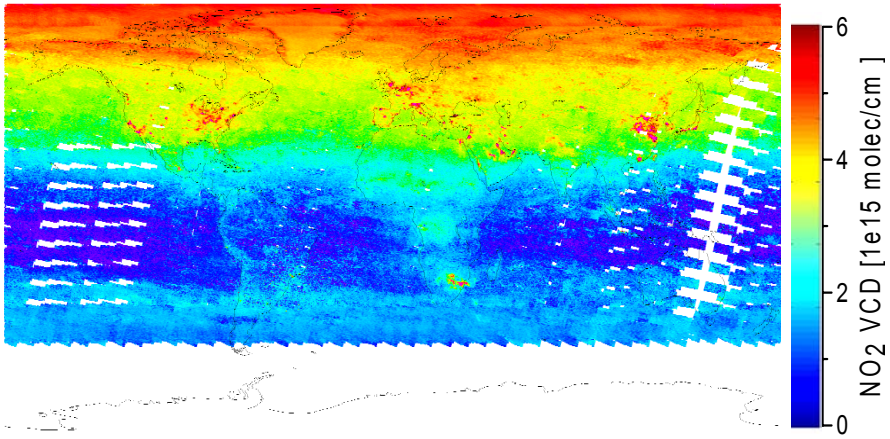

Fig. 28 Monthly mean of the total NO_2 VCD analysed from SCIAMACHY for June 2003 (scientific product developed at University of Heidelberg, Beirle et al. [2004]). During Summer high stratospheric values are found in high northern latitudes.

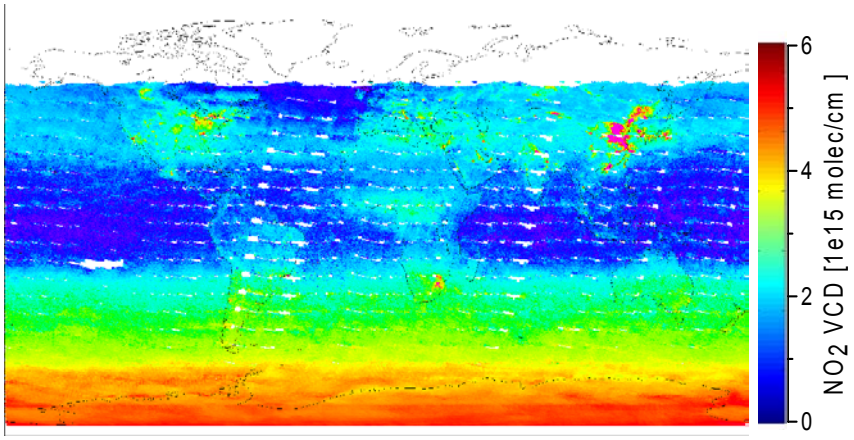

Fig. 29 Monthly mean of the total NO_2 VCD analysed from SCIAMACHY for December 2003 (scientific product developed at University of Heidelberg, Beirle et al. [2004]). During Winter high stratospheric values are found in high southern latitudes.

In Figures 30 – 33 the NO_2 VCDs measured at Kiruna are compared to those from SCIAMACHY (averages or minima within 200km, scientific product of the University of Bremen). In general, a very good agreement is found; in particular the seasonal cycle is well present in both data sets. If one looks in some more detail (Figures 31 – 32) it can be seen that the minima fit much better to the ground based data. This finding indicates that probably remaining (small) NO_2 concentrations are present over the station. This is not surprising, as the measurement site is close to the city of Kiruna, and its airport.

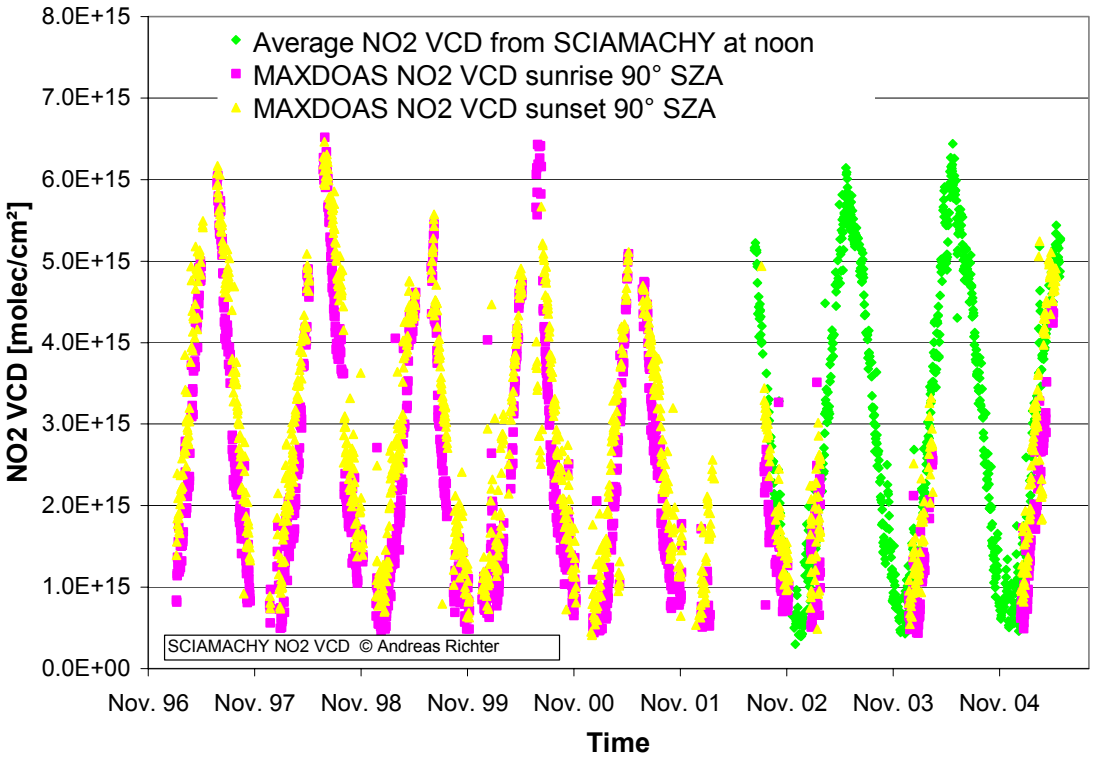

Fig. 30 Time series of NO_2 VCDs measured by the Kiruna instrument since December 1996. From 2002 to 2005 also the time series of average SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) are shown.

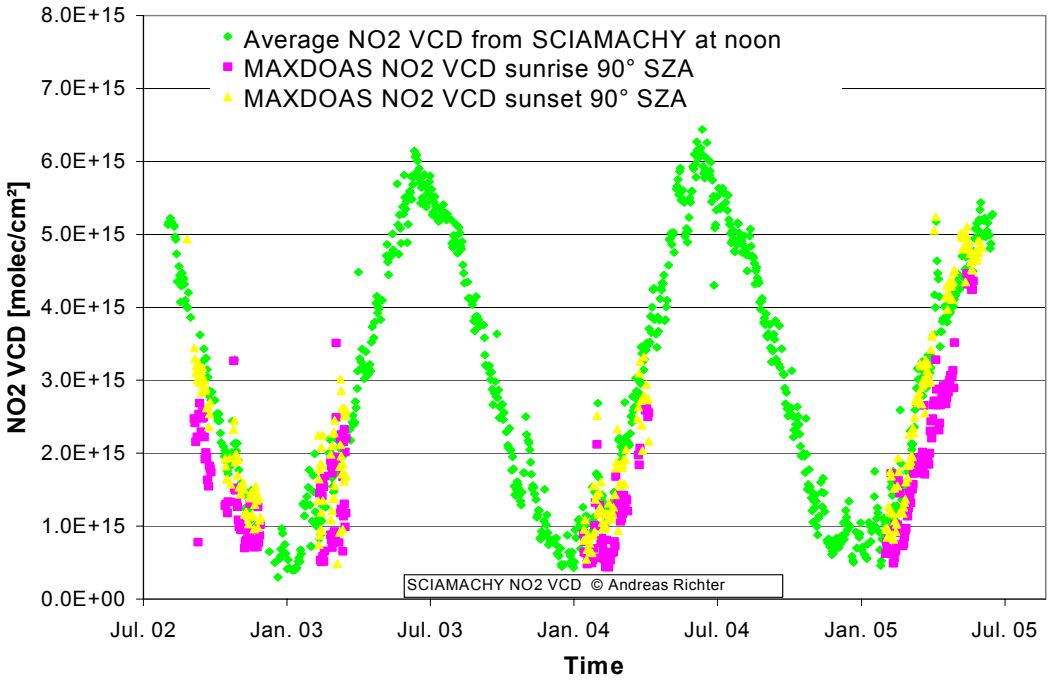

Fig. 31 Time series of NO_2 VCDs measured by the Kiruna instrument and average SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) for the period 2002 – 2005.

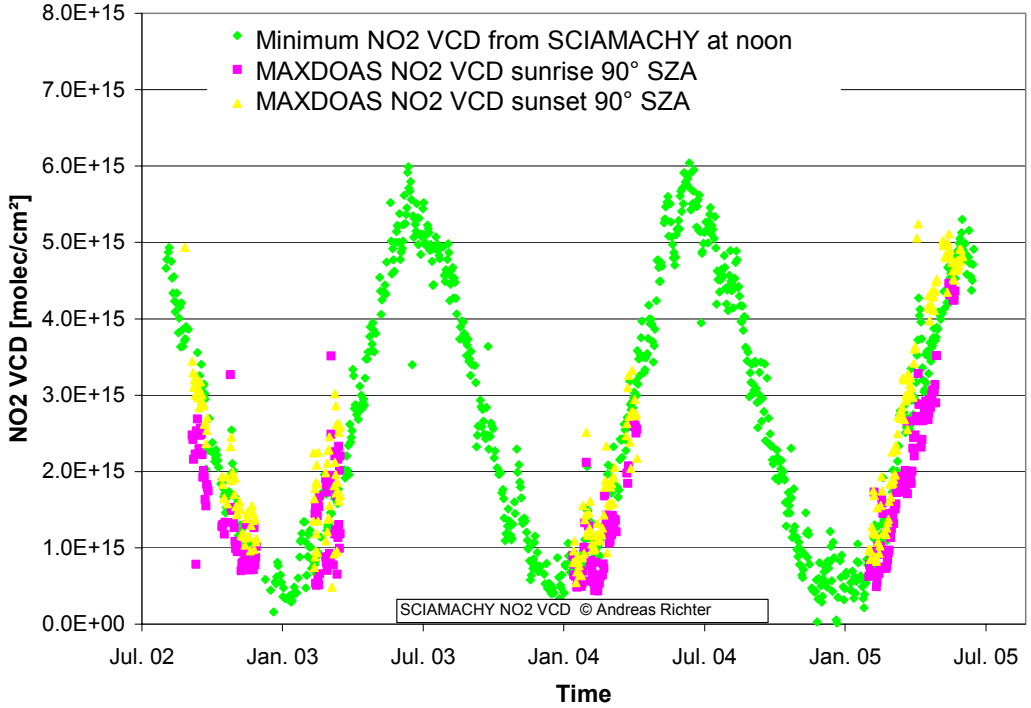

Fig. 32 Time series of NO_2 VCDs measured by the Kiruna instrument and minimum SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) for the period 2002 – 2005. The minimum SCIAMACHY NO_2 VCDs fit better than the average values (see Fig. 31) indicating the presence of tropospheric NO_2.

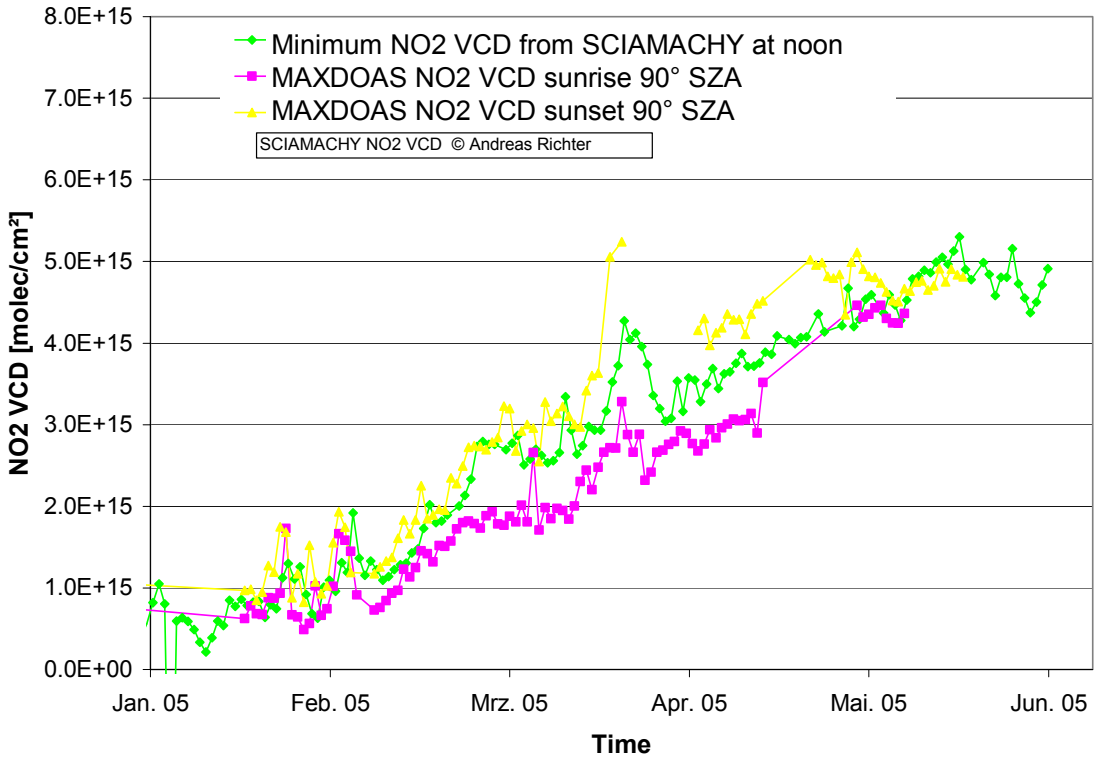

Fig. 33 Time series of NO_2 VCDs measured by the Kiruna instrument and minimum SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) for the first half of 2005. The SCIAMACHY NO_2 VCDs are between the Kiruna AM and PM data probably indicating remaining low NO_2 contributions from the troposphere.

In Figures 34 – 35 the NO_2 VCDs measured at Paramaribo are compared to those from SCIAMACHY (averages or minima within 200km, scientific product of the University of Bremen). In general, a very good agreement is found; in particular the seasonal cycle (although very weak in the Tropics) is well present in both data sets. If one looks in some more detail it can be seen that the minima fit much better to the ground based data. This finding indicates that probably remaining (substantial) NO_2 concentrations are present over the station. This is not surprising, as the measurement site is close to the city of Pramaribo.

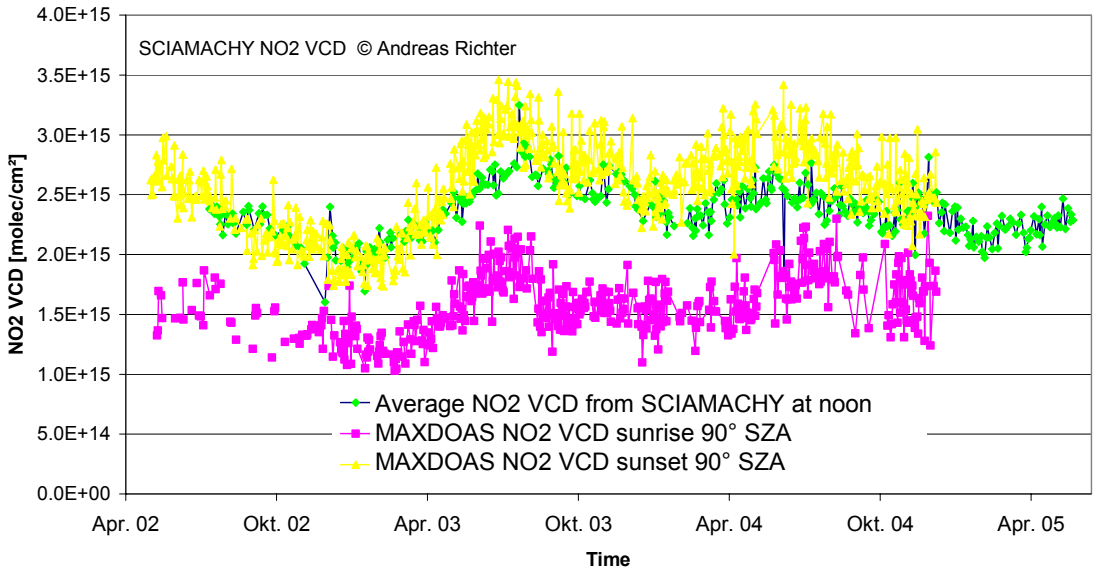

Fig. 34 Time series of NO_2 VCDs measured by the Paramaribo instrument and average SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) for the period 2002 – 2005.

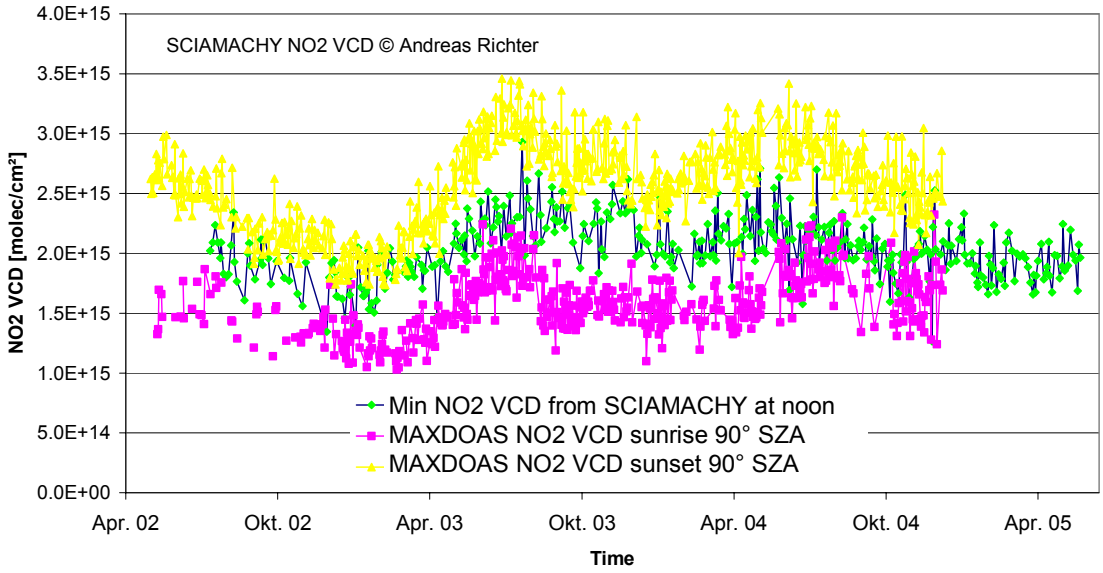

Fig. 35 Time series of NO_2 VCDs measured by the Paramaribo instrument and minimum SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) for the period 2002 – 2005.

In Figures 36 – 38 the NO_2 VCDs measured at Neumayer are compared to those from SCIAMACHY (averages or minima within 200km, scientific product of the University of Bremen). In general, a very good agreement is found; in particular the seasonal cycle is well present in both data sets. In contrast to the measurements at Kiruna and Pramaribo, no indications for significant amounts of tropospheric NO_2 were found.

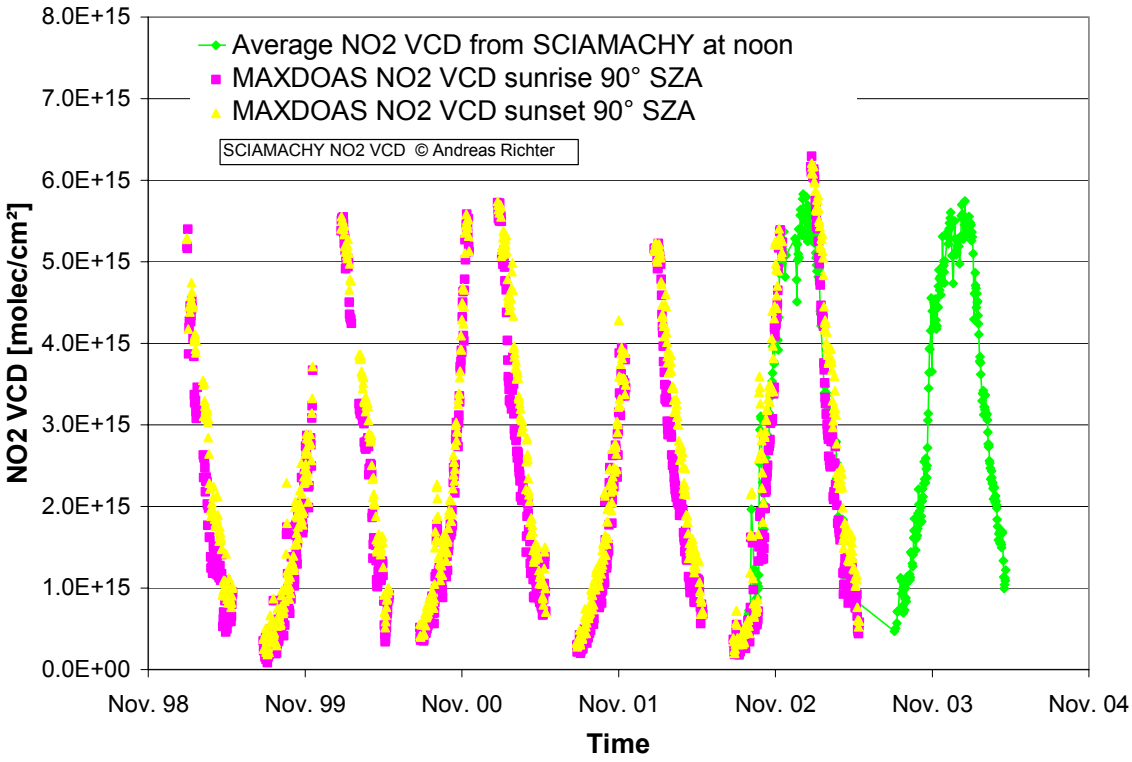

Fig. 36 Time series of NO_2 VCDs measured by the Neumayer instrument since January 1999. From 2002 to 2005 also the time series of average SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen) are shown.

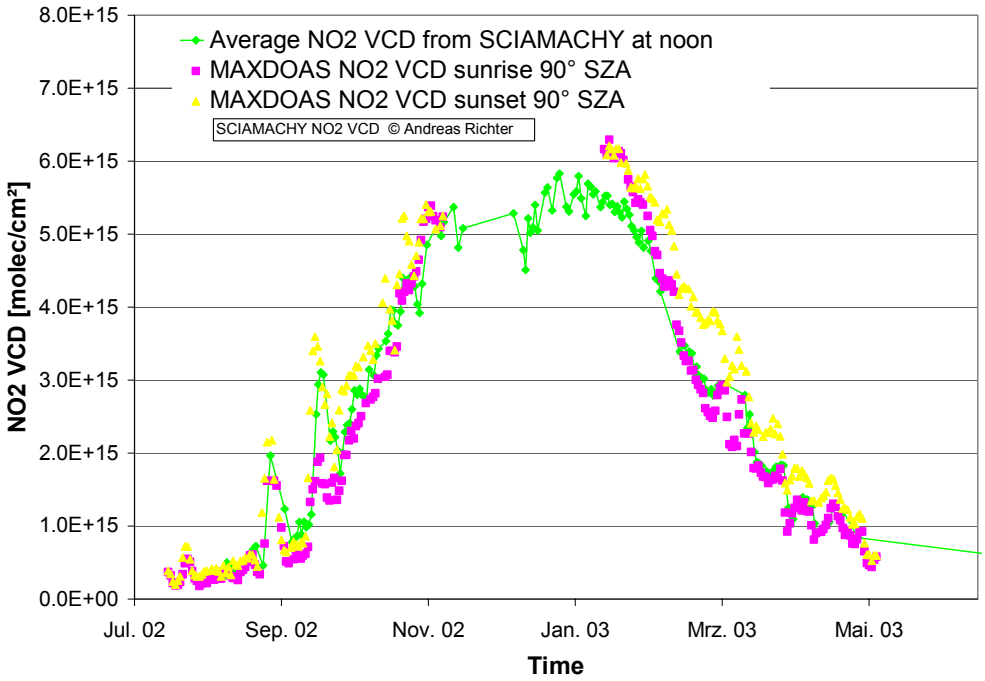

Fig. 37 Time series of NO_2 VCDs measured by the Neumayer instrument for summer 2002 to summer 2003 together with the average SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen).

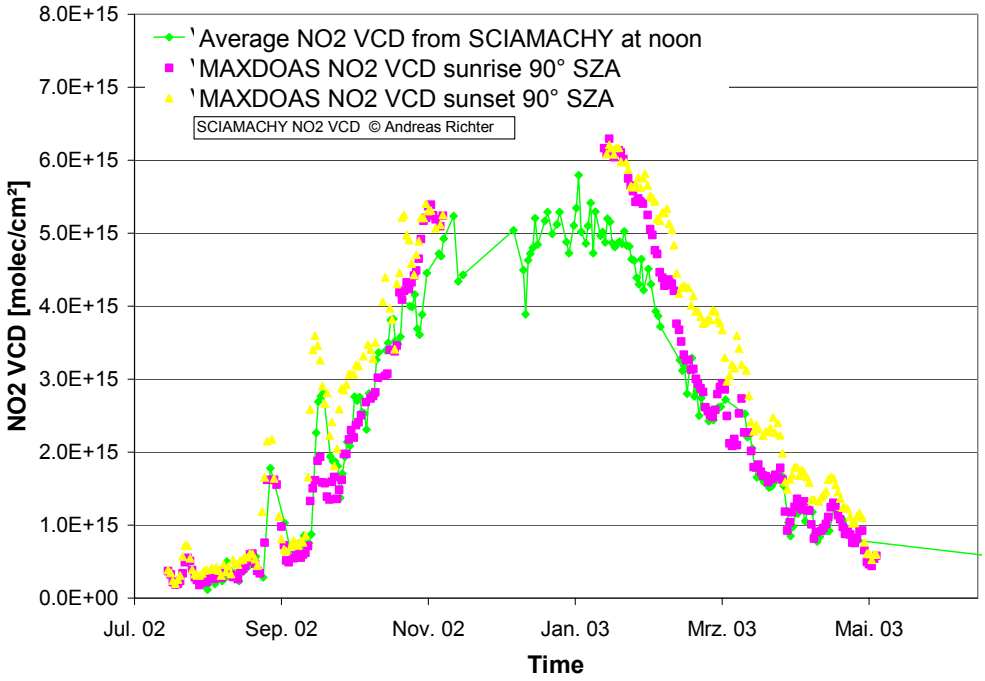

Fig. 38 Time series of NO_2 VCDs measured by the Neumayer instrument for summer 2002 to summer 2003 together with the minimum SCIAMACHY NO_2 VCDs (within 200km, scientific product of the University of Bremen).

2.5.3 Tropospheric NO_2

So far, no official tropospheric NO_2 data product derived from SCIAMACHY is available. Also, the scientific products are not retrieved under standardised conditions. Thus, currently a validation of tropospheric SCIAMACHY products is not possible. Nevertheless, our data sets of tropospheric NO_2 columns can serve for the validation of future developments of tropospheric NO_2 satellite products.

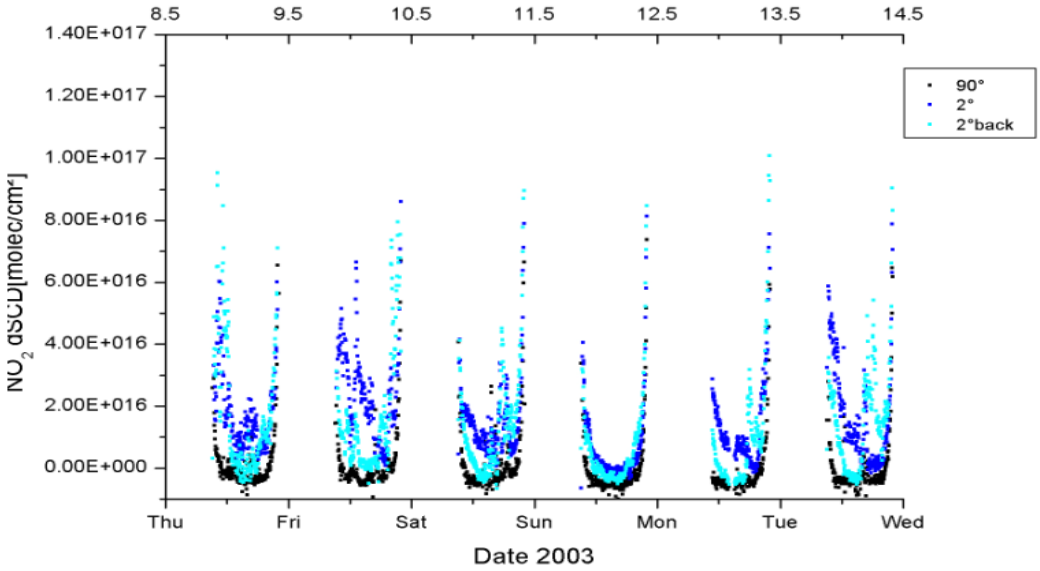

Fig. 39 NO_2 SCDs analysed for the zenith telescope and for two telescopes with 2° elevation of the Suriname instrument for six days. The enhanced NO_2 SCDs for the low elevation angles indicate substantial tropospheric NO_2 concentrations.

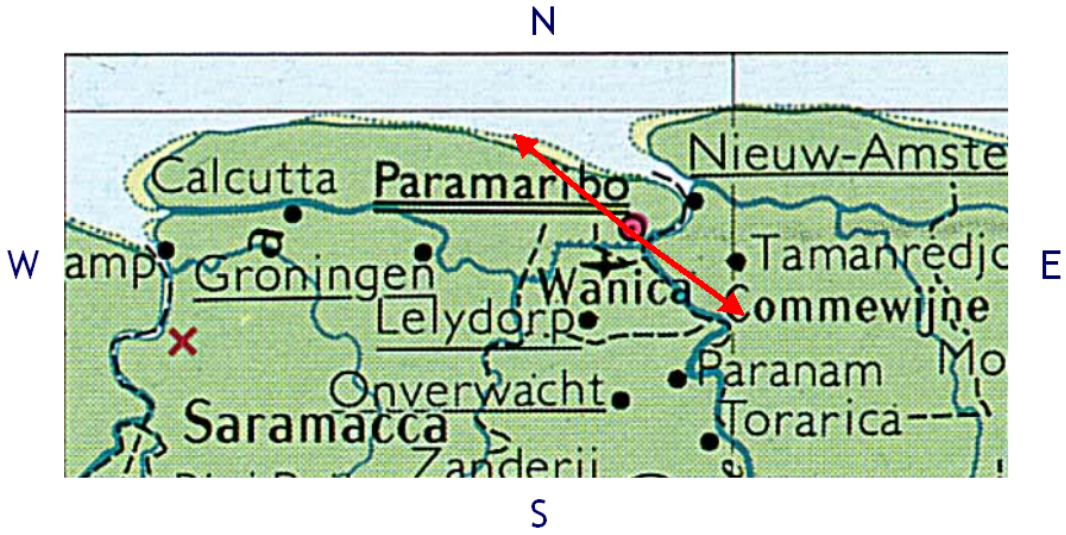

Fig. 40 Orientation of the two telescopes with 2° elevation, but opposite azimuth angles.

According to the MAXDOAS principle, tropospheric columns can be derived from the differences between the different elevation angles. While at Kiruna and especially

at Neumayer, the tropospheric contributions to the atmospheric total column of NO_2 are small (see above), at Paramaribo substantial tropospheric NO_2 concentrations are typically present. In Figures 39-41 examples for the measurements at Suriname are given. In Fig. 39 the NO_2 SCDs analysed for the zenith telescope and for two telescopes with 2° elevation are shown for six days. Several interesting findings can be seen:

- as expected the NO_2 SCDs for the low elevation angles are typically smaller than those of the zenith telescopes. This indicates significant amounts of NO_2 in the troposphere.
- the NO_2 SCDs for the zenith telescopes show a u-shaped diurnal variation which is typical for the stratosphere.
- the two measurements for 2° elevation are pointing in opposite azimuth directions (see Fig. 40). The respective NO_2 SCDs show a different temporal evolution indicating a NO_2 concentration field with strong horizontal gradients moving over the measurement site. This finding shows the potential for the identification of spatial inhomogneities within a satellite ground pixel.
- During Sunday the NO_2 SCDs are much smaller than during working days, indicating reduced NO_x emissions.

Since the measurements for 2° elevation and zenith direction have similar sensitivity for the stratosphere, but different sensitivity for the troposphere; the difference contains solely the tropospheric signal (see Fig. 41).

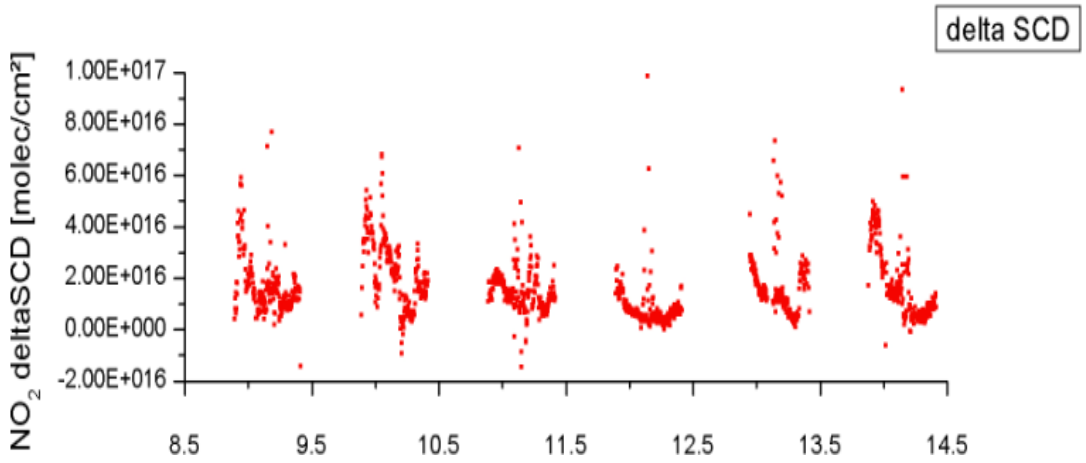

Fig. 41 Difference of the NO_2 SCDs of the telescope at 2° elevation and zenith direction. Both measurements have similar sensitivity for the stratosphere, but different sensitivity for the troposphere; the difference contains solely the tropospheric signal.

2.5.4 Stratospheric BrO

Because of the strong photochemical variability of BrO, the validation of the total BrO VCD is a major challenge. First, the satellite observations of BrO were taken around local noon, while typically the ground based BrO VCDs are retrieved for 90° SZA.

In addition, for the ground based measurements themselves, it is difficult to determine the absorption of the Fraunhofer reference spectrum with the method of Langley plot, because the BrO VCD varies during the day.
Both problems can only be adequately dealt with if photochemical modelling is included, which is beyond the scope of this project. Nevertheless, even with the above limitations already important information concerning the order of magnitude and the seasonal dependence of the satellite data can be derived.
In Fig. 42 global mean BrO VCD derived from SCIAMACHY for March 2003 is shown (data are from IASB/BIRA, Brussels). The overall distribution with high values at the poles and low values at the tropics is caused by the stratosperic column; the areas with high values in the Arctic are caused by enhanced tropospheric concentrations.
In Fig. 43 the BrO VCDs measured from the Kiruna ground based instrument are compared to those from SCIAMACHY and GOME (scientific data products from IASB/BIRA, Brussels, http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). In general, all three data sets show a good agreement, in particular the same seasonal variation. The ground based data are, however, systematically smaller than the satellite measurements, which is in agreement with the expectations from photochemistry.
Fig. 44 displays the BrO VCDs measured by the ground based instrument at Paramaribo. As expected from stratospheric photochemistry, the values are small and show no pronounced seasonal variation. They are in good agreement with the tropical SCIAMACHY BrO VCDs as shown in Fig. 42.

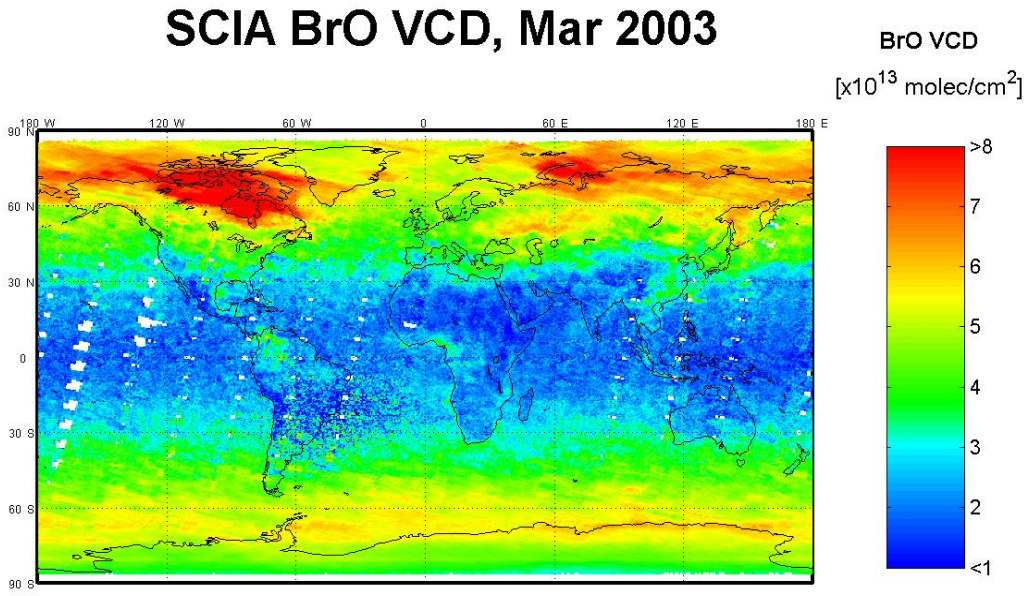

Fig. 42 Global mean BrO VCD derived from SCIAMACHY for March 2003 (data are from IASB/BIRA, Brussels, see http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). The overall distribution with high values at the poles and low values at the tropics is caused by the stratosperic column; the areas with high values in the Arctic are caused by enhanced tropospheric concentrations.

In Figures 45 and 46 the BrO DSCDs measured from the ground based instruments at Neumayer and Arrival Heights are compared to the BrO SCDs analysed from SCIAMACHY and GOME (scientific data products from IASB/BIRA, Brussels, see http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). Here, so far no BrO VCDs could be derived from the BrO DSCDs; thus from this comparison only rough qualitative conclusions can be drawn. Nevertheless, the order of magnitude and the seasonal variation is in good agreement for all three data sets at both stations.

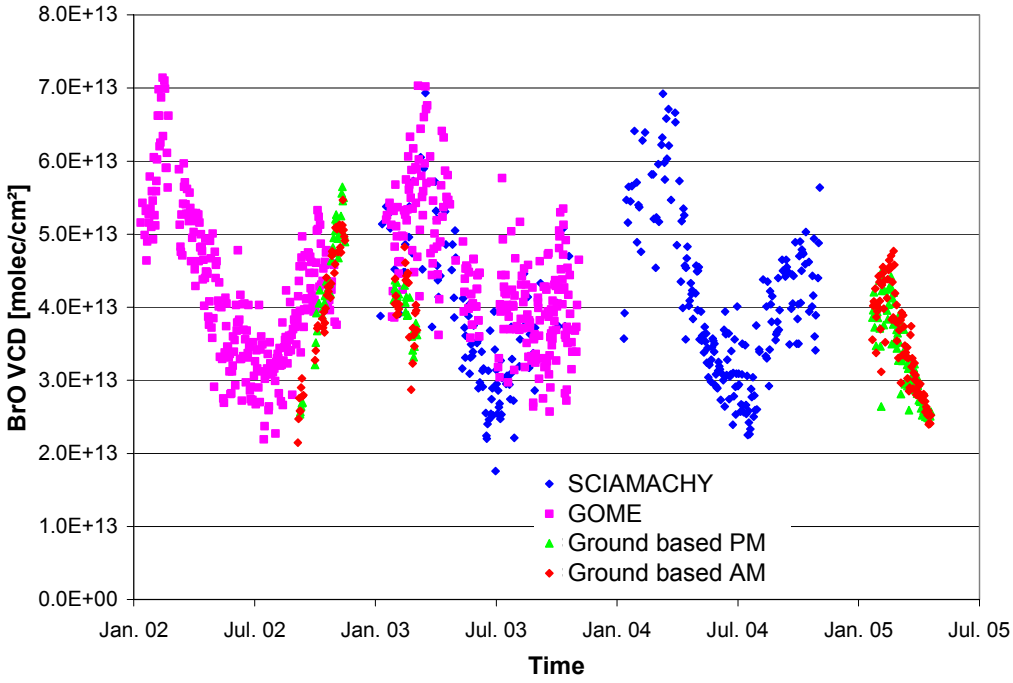

Fig. 43 Comparison of the BrO VCDs measured from the Kiruna ground based instrument with those from SCIAMACHY and GOME (scientific data products from IASB/BIRA, Brussels, see http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). In general, all three data sets show a good agreement, in particular the same seasonal variation. The ground based data are systematically smaller than the satellite measurements, which is in agreement with the expectations from photochemistry.

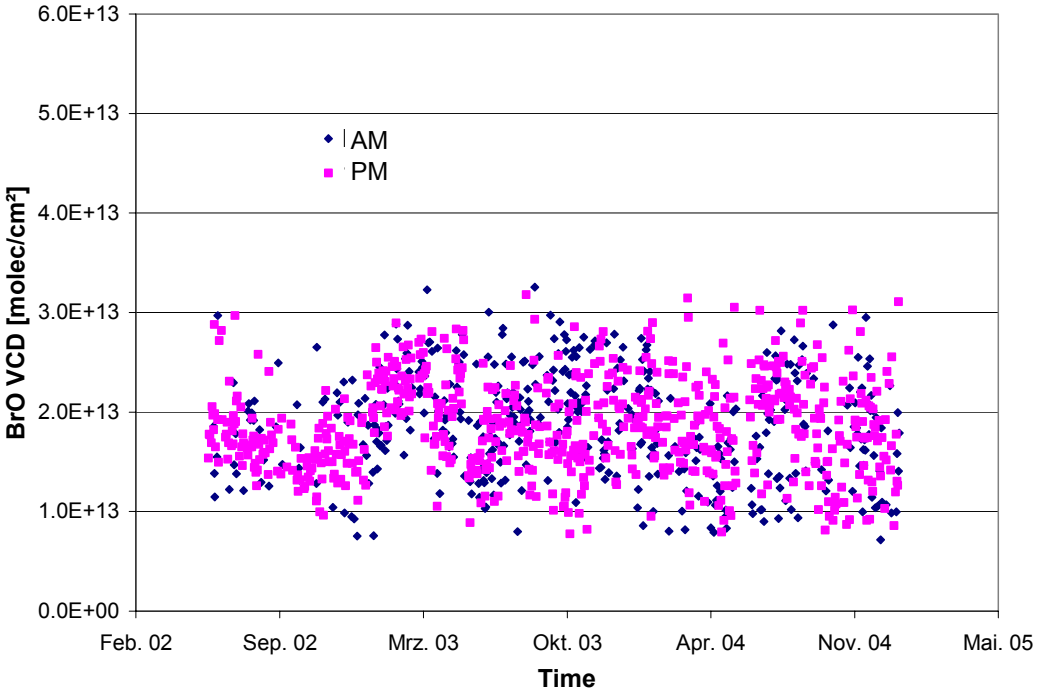

Fig. 44 BrO VCDs measured from the Paramaribo ground based instrument. In the tropics, the BrO VCDs show only a week seasonal variation. The ground based data are in good agreement with the SCIAMACHY data shown in Fig. 42.

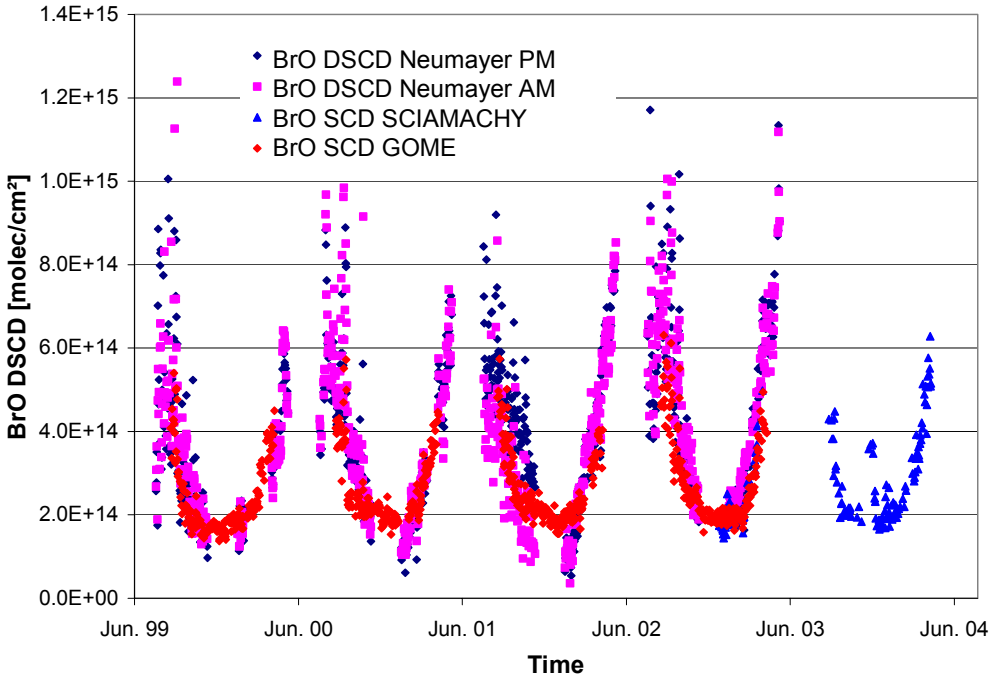

Fig. 45 Comparison of the BrO DSCDs measured from the Neumayer ground based instrument with the BrO SCDs analysed from SCIAMACHY and GOME (scientific data products from IASB/BIRA, Brussels, see http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). Although all three data sets show a good qualitative agreement, no quantitative conclusions can be drawn from this comparison, because no conversion to BrO VCDs has been performed.

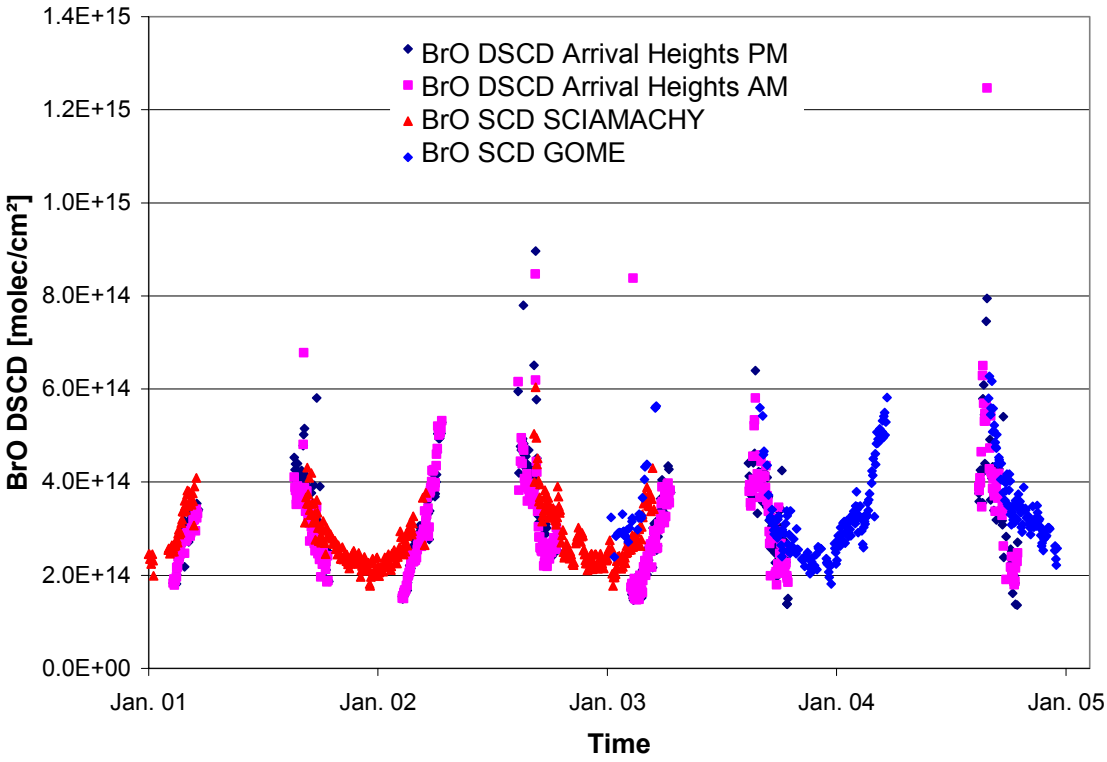

Fig. 46 Comparison of the BrO DSCDs measured from the Arrival Heights ground based instrument with the BrO SCDs analysed from SCIAMACHY and GOME (scientific data products from IASB/BIRA, Brussels, see http://www.oma.be/BIRA-IASB/Molecules/BrO/intro_GOME_SCIA.php). Although all three data sets show a good qualitative agreement, no quantitative conclusions can be drawn from this comparison, because no conversion to BrO VCDs has been performed.

2.5.5 Stratospheric OClO

Similarly to BrO, also the comparison of satellite and ground based data for OClO is not straight forward because of the strong photochemical variability of stratospheric OClO. In particular, so far no official SCIAMACHY OClO data product is available for validation so far. Nevertheless, the OClO DSCDs sets analysed from the ground based stations of Kiruna and Neumayer provide a very well suited data set for future validation studies.

In Fig. 47 the OClO DSCDs for the ground based measurements at Kiruna are shown. As expected for the northern polar stratosphere, the OClO DSCDs are different for different winters. Strong chlorine activation can only occur if stratospheric temperatures allow the formation of polar stratospheric clouds. Accordingly, enhanced OClO DSCDs are found for the cold winters 1999/2000 and 2004/2005. During these winters also strong ozone destruction was observed (see Fig. 48).

In contrast to the northern hemisphere, the stratospheric temperatures over Antarctica are very similar for different years. Accordingly, similar OClO DSCDs are observed

for the various winters (Fig. 49). The values are systematically higher compared to those over Kiruna.

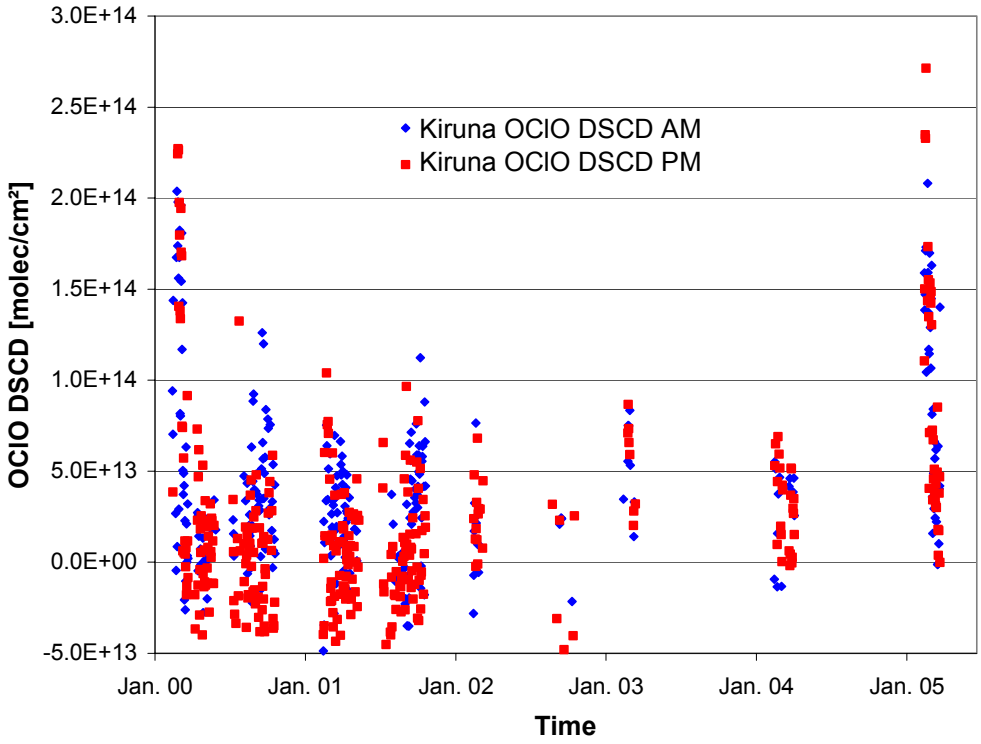

Fig. 47 OClO DSCDs over Kiruna for different polar winters. High values were detected for the cold winters 1999/2000 and 2004/2005.

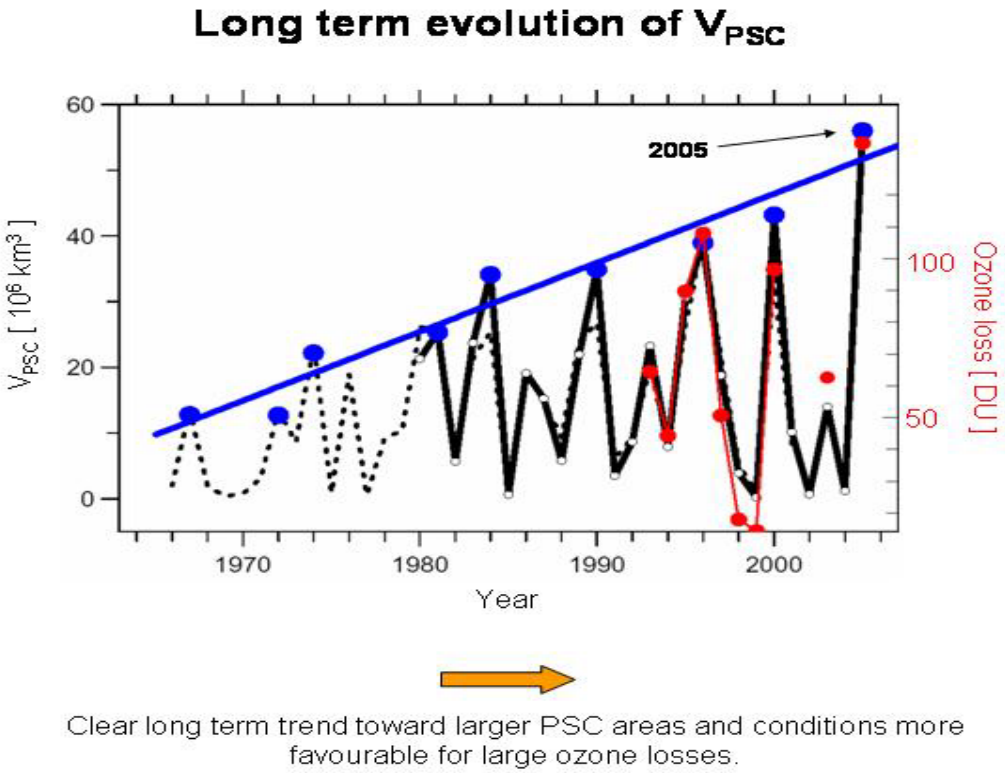

Fig. 48 Volume of polar stratospheric clouds and Ozone loss for different Antarctic winters (© Markus Rex, see http://www.realclimate.org/). Strong ozone destruction

was observed during the winters with high chlorine activation indicated by high OClO DSCDs over Kiruna (see Fig. 47).

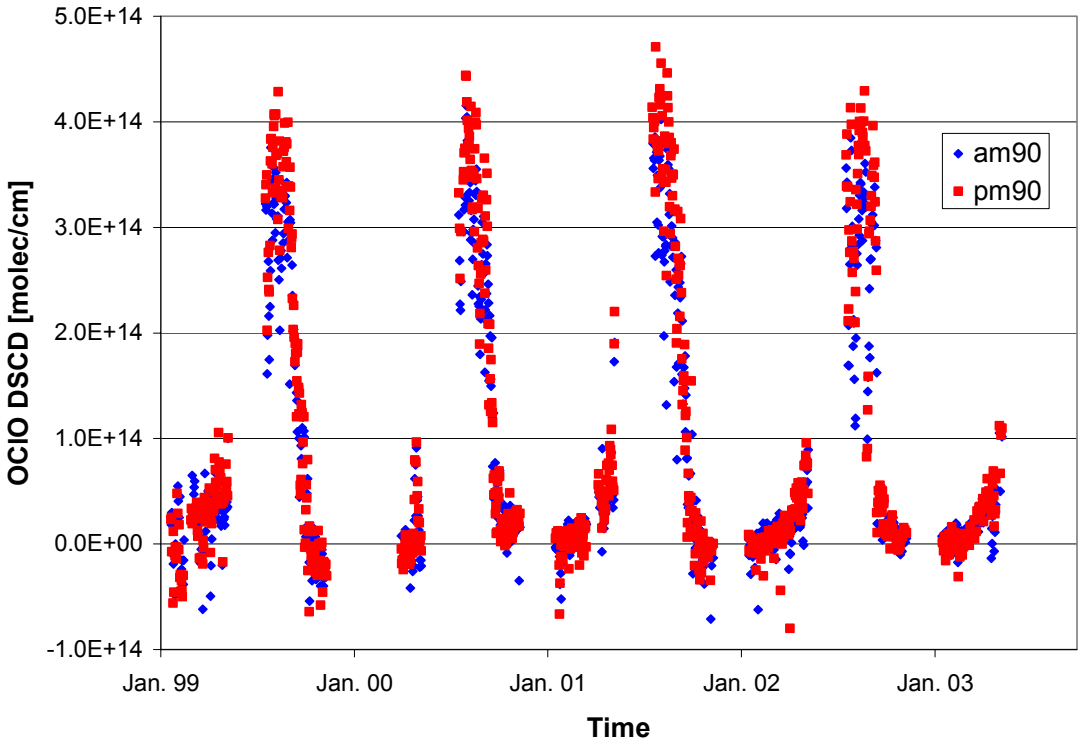

Fig. 49 OClO DSCDs over Neumayer for different polar winters. Similar values are found for all years.

2.5.6 Tropospheric H_2O and O_4

Besides the 'classical' validation trace gases (like NO_2 or BrO), also the tropospheric columns of H_2O and O_4 can be retrieved from MAXDOAS observations.
So far, no official SCIAMACHY data products for H_2O and O_4 are available. Thus, currently a validation of tropospheric SCIAMACHY products is not possible. Nevertheless, our data sets of tropospheric H_2O and O_4 columns can serve for the validation of future developments of tropospheric H_2O satellite products.
Especially from measurements of the oxygen dimer O_4 information on the radiative transfer, in particular on the presence of clouds, can be retrieved. In contrast to H_2O, the atmospheric concentration of O_4 is almost constant. Thus variations in the measured O_4 absorption can be almost exclusively attributed to light path variations.
This makes O_4 observations a very important tool for the correct interpretation of tropospheric trace gas observations [Wagner and Platt, 1998; Wagner et al., 1998; Wagner et al., 2002; Wagner et al., 2004; Sinreich et al., 2005].
In Fig. 50 the H_2O DSCDs for the different elevation angles of the Paramaribo MAXDOAS instrument are shown. The Highest values are found for the lowest

elevation angles indicating that the bulk of the H_2O column is located very close to the ground.
Similar results are also found for O_4 (Fig. 51). However, the DSCDs for the telescopes at 2° and 5° are very similar indicating that the O_4 profile is not as closely located to the ground as the H_2O profile. The scatter of the data indicates the presence of clouds.

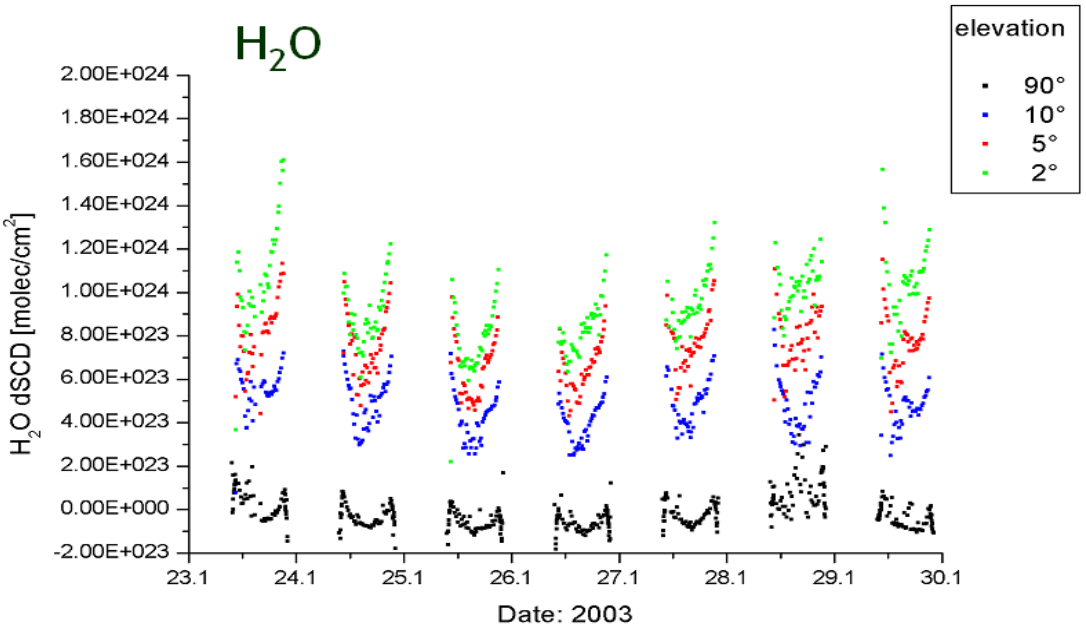

Fig. 50 H_2O DSCDs for the different elevation angles of the Paramaribo MAXDOAS instrument. The Highest values are found for the lowest elevation angles indicating that the bulk of the H_2O column is located very close to the ground.

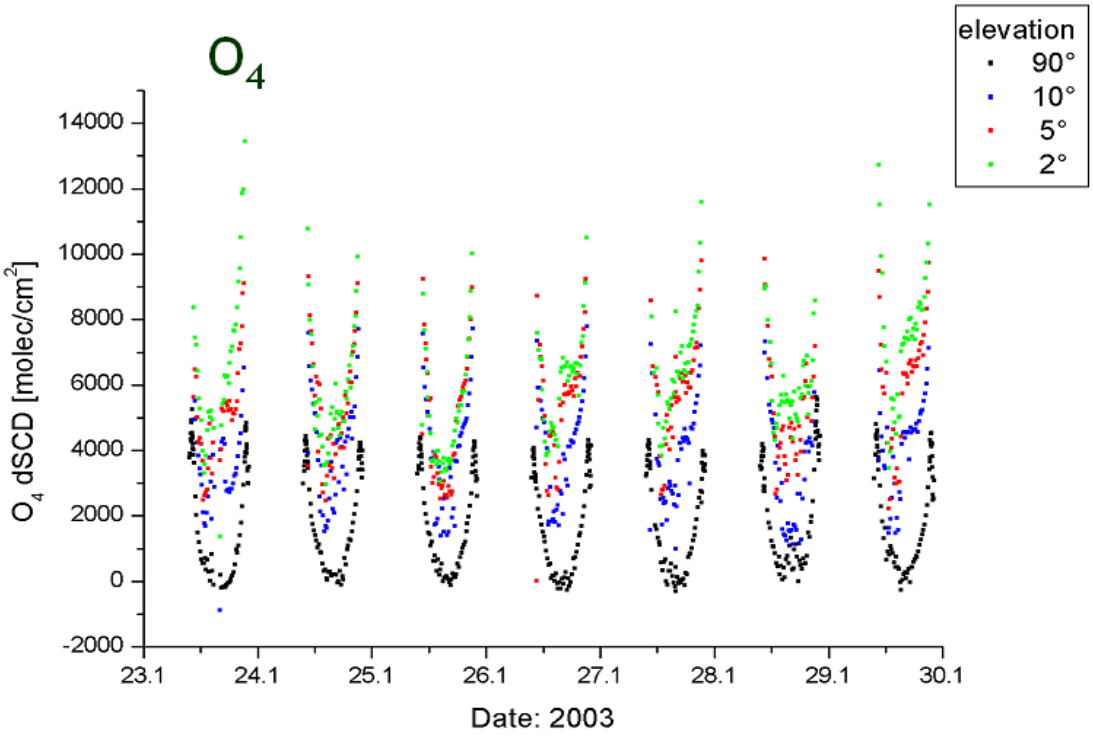

Fig. 51 O_4 DSCDs for the different elevation angles of the Paramaribo MAXDOAS instrument. In contrast to H_2O, the DSCDs for the telescopes at 2° and 5° are very similar indicating that the O_4 profile is not as closely located to the ground as the H_2O profile. The scatter of the data indicates the presence of clouds.

2.6 Zusammenfassung

In Rahmen des Projektes wurde ein MAXDOAS-Instrument entwickelt und auf der Meteorologischen Station von Paramaribo (Surinam) installiert (http://www.knmi.nl/samenw/star/station.html). Das Instrument führt seit May 2002 kontinuierlich Messungen durch.
Zusätzlich zu dem neuen Instrument in den Tropen wurden die Daten der bereits bestehenden Instrumente der Stationen Kiruna (Schweden, http://www.irf.se/program/afp/doas/) sowie Neumayer und Arrival Heights (beide Antarktis) zur Validation der Satellitenmessungen herangezogen. Zu diesem Zweck wurden die Instrumente kontinuierlich betreut und falls notwendig auch vor Ort repariert. Zudem wurden Veränderungen an den bestehenden Instrumenten durchgeführt: in Kiruna wurde der vis-Spektrometer im Mai 2002 durch einen modernen Ocean-Optics-Spektrographen ersetzt. Anfang 2003 wurde für das Neumeyer-Gerät ein neues MAXDOAS-Teleskop installiert.
Die Datenanalyse wurde weiter verbessert und die Langzeitdatensätze nach dem neusten Stand der Auswertetechnik konsistent analysiert.
Aus den gewonnenen Meßdaten wurden die folgenden Spurenstoffprodukte analysiert:

- stratosphärische und troposphärische Säulendichten von NO_2
- stratosphärische Säulendichten von O_3
- stratosphärische Säulendichten von BrO
- stratosphärische Säulendichten von OClO
- troposphärische Säulendichten von H_2O
- troposphärische Säulendichten von O_4

Derzeit ist der Umfang an SCIAMACHY-Daten zur Validation noch sehr begrenzt. In diesem Projekt wurde die Validierung daher noch auf die stratosphärischen Säulendichten von O_3, NO_2 und BrO beschränkt.
Sowohl für O_3 und NO_2 wurde eine sehr gute Übereinstimmung zwischen den Bodenmessungen und den Satellitendaten gefunden. Verbleibende Abweichungen sind im Fall von NO_2 sehr wahrscheinlich weitgehend auf troposphärsche Beiträge zurückzuführen, da sich hier die Empfindlichkeit von Satelliten- und Bodeninstrumenten sehr unterscheidet. Zusätzlich könnte auch die Variabilität der stratosphärischen Chemie Abweichungen begründen.
Speziell die gute Übereinstimmung für die tropische Station in Paramaribo zeigt die hohe Qualität der NO_2-Messungen von SCIAMACHY. Sowohl der Absorptionsweg als auch die stratosphärischen NO_2-Konzentrationen selbst sind nur klein in den Tropen, was zu einem geringen Signal-Rausch-Verhältnis und zu relativ hohen (potentiellen) systematischen Fehlern führt. Die gute Übereinstimmung mit den Bodendaten in den Tropen zeigt, daß diese prinzipiellen Schwierigketen das NO_2 Produkt von SCIAMACHY nicht signifikant beeinträchtigen.
Auch für stratosphärisches BrO wurde für alle Stationen eine gute Übereinstimmung gefunden. Allerdings erlauben die bisherigen Vergleiche nur qualitative Aussagen, da

für einen detaillierten Vergleich der Einfluß der stratosphärischen Photochmie anhand von Modelldaten berücksichtigt werden muß.
Die troposphärischen Spurenstoffdaten der MAXDOAS-Bodenmessungen stellen einen wertvollen Datensatz für die Validierung zukünftiger Satellitendaten dar.

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3 Erfolgskontrollbericht

3.1 Beitrag des Ergebnisses zu den förderpolitischen Zielen

Die Validation von SCIAMACHY-Daten mit bodengebundenen Messungen ist ein wesentlicher Aspekts der Aufgaben, deren Bedeutung in den Dokumenten ‘Validation Requirements Document’ und ‘SCIAMACHY Core Validation’ beschrieben ist. Eine umfassende und genaue Validierung ist eine entscheidende Vorraussetzung für die Akzeptanz und Nutzung der Satellitendaten durch die wissenschaftliche Gemeinschaft. Speziell die Messungen von SCIAMACHY auf ENVISAT tragen entscheidend zur Erfassung der globalen Spurenstoffverteilung und damit zum Verständnis von Phänomenen wie Luftverschmutzung und Klimawandel bei.

3.2 Wissenschaftlich-technisches Ergebnis des Vorhabens

Die wissenschaftlichen und technischen Ergebnisse des Vorhabens wurden im Abschnitt 2 beschrieben.

3.3 Fortschreibung des Verwertungsplans

Es wurden weder Erfindungen gemacht, noch Schutzrechte in Anspruch genommen. Da es sich um reine Grundlagenforschung handelt, entsprechen wirtschaftliche Erfolgsaussichten nicht der Zielsetzung des Projektes.
Die Daten werden in Kürze auf die CALVAL-Datenbank am NILU übertragen.

3.4 Arbeiten, die zu keiner Lösung geführt haben

Wie im Abschnitt 2 ausführlich erläutert, waren die bodengestützen-Messungen sehr erfolgreich. Neben den erzeugten einmaligen Validationsdatensätzen wurden insbesondere auf technologischer und algorithmischer Seite deutliche Fortschritte erzielt.
Während die Instrumnte der Stationen Paramaribo, Neumayer und Arrival Heights sehr stabil arbeiteten, kam es beim Instrument in Kiruna leider zu einigen Meßausfällen. Diese waren hauptsächlich durch Computerausfälle begründet. Obwohl es sich meist nur um den Ausfall von Standard-Komponenten handelte, war ein Ersatz schwierig, weil die Instrumente in Kiruna schon 15 Jahre alt sind und daher die Kompatibiltät der Komponenten problematisch war. Glücklicherweise ist es bisher immer wieder gelungen, die Geräte im operationellen Betrieb neu zu starten. Dies konnte teilweise durch Ferndiagnose aus Heidelberg, zum Teil aber nur durch Reparaturbesuche vor Ort erreicht werden. Dennoch muß dieser Aufwand als gerechtfertigt angesehen werden, da die DOAS-Messungen in Kiruna eine der längsten Meßreihen in der Arktis darstellen. Ein Ersatz des Gerätes (oder von wichtien Instrumentkomponenten) würde die Kontinuität dieser nun fast 10-jährigen Meßreihe unterbrechen.

3.5 Präsentationsmöglichkeiten für mögliche Nutzer

Ein großer Teil der Ergebnisse des Projektes wurde bereits in rezensierten Artikeln sowie in Konferenzbeiträgen veröffentlicht. Weitere Artikel sind zur Zeit in Vorbereitung.

3.6 Einhaltung der Ausgaben- und Zeitplanung

Die Ausgabenplanung wurde eingehalten. Die Zeitplanung mußte aufgrund der Startverschiebungen von ENVISAT und der Verzögerungen bei der Bereitstellung der operationellen Datenprodukte überarbeitet werden.

3.7 Fortschreibung des Verwertungsplans

Die gewonnen Daten stehen auch nach Ende des Projektes noch zur Verfügung, und können zur Validation von neuen Versionen der operationellen Datenprodukte verwendet werden. Die Ergebnisse werden in Kürze in die NILU Datenbank übertragen. Bei künftigen Prozessor-Verbesserungen stehen sowohl die Daten, als auch der Projektleiter zur Unterstützung von Vergleichen zur Verfügung.

3.7.1 Erfindungen und Patentanmeldungen

Erfindungen und Patentanmeldungen erfolgten nicht.

3.7.2 Wirtschaftlichen Erfolgaussichten nach Projektende

Da es sich bei dem Projekt um reine Grundlagenforschung handelt, gibt es keine wirtschaftlichen Erfolgaussichten nach Projektende.

3.7.3 Wissenschaftliche und / oder technische Erfolgsaussichten nach Projektende

Die im Projekt gewonnenen Ergebnisse lassen sich in einer Reihe von wissenschaftlichen Anwendungen nutzen:

• Weitere Validation von SCIAMACHY Produkten
• Fortführung der MAXDOAS-Messungen
• Anwendung der entwickelten Algorithmen auf andere MAXDOAS Messungen
• Weiterentwicklung und Anwendung tomographischer Inversionsmethoden zur Analyse der atmosphärischen Spurenstoffverteilung
• Anwendung der Meßdaten für atmosphärische Studien

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